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Interim process report for the safety assessment SR-Can

Svensk Kärnbränslehantering AB

August 2004

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Preface

This report gives an interim account of how processes, important for the long-term evolution of a KBS-3 repository for spent nuclear fuel, will be documented in the safety assessment SR-Can. Buffer processes are used to exemplify the documentation.

Most of the material in the report was contributed by selected experts in the field, in particular Ola Karnland and Lennart Börgesson, Clay Technology AB. See section 1.2 for a complete list of contributors. The sub-sections describing the handling in the safety assessment of the various processes were written by Patrik Sellin and Allan Hedin, SKB in collaboration with the concerned experts. Patrik Sellin has edited the report.

Stockholm, August 2004

Allan Hedin
Project leader, SR-Can

Contents

1 Introduction

This report is a documentation of buffer processes identified as relevant to the long-term safety of a KBS-3 repository. The report is part of the interim reporting of the safety assessment SR-Can, see further the Interim main report, /SKB, 2004a/. The final SR-Can reporting will support SKB's application to build an Encapsulation plant for spent nuclear fuel and is to be produced in 2006.

The purpose of this report is to document the scientific knowledge of the processes to a level required for an adequate treatment in the safety assessment. The documentation is thus from a scientific point of not exhaustive since such a treatment is neither necessary for the purposes of the safety assessment nor possible within the scope of an assessment.

The purpose is further to determine a handling of each process in the safety assessment and to demonstrate how uncertainties are taken care of, given the suggested handling.

The process documentations in the SR 97 version of the Process report /SKB, 1999/ is a starting point for this SR-Can interim version. As further described in the Interim main report, the list of relevant processes has been reviewed and slightly extended by comparison to other databases. Furthermore, the backfill has been included as a system part of its own, rather than being described together with the buffer as in SR 97.

Apart from giving an interim account of the documentation and handling of buffer processes in SR-Can, this report is meant to serve as a template for the forthcoming documentation of processes occurring in other parts of the repository system. A complete list of processes can be found in the Interim FEP report for the safety assessment SR-Can /SKB, 2004b/.

All material presented in this document is preliminary in nature and will possibly be updated as the SR-Can project progresses.

1.1 Structure for process descriptions

All identified processes are documented using the following template, where many of the headings are the same as those used in the SR 97 report:

Overview/general description

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

A table is produced, documenting how the process is influenced by the specified set of physical variables in the relevant system component and how the process influences the variables.

Boundary conditions

The boundary conditions for each process will be discussed. These refer to the boundaries of the relevant system part. For example, for buffer processes the boundaries are the buffer interfaces with the canister, the walls of the deposition hole and the backfill. The processes for which boundary conditions need to be described are, in general, related to transport of material or energy across the boundaries. For example for chemical processes occurring within a system component, like illitisation in the buffer, the discussion of boundary conditions will relate to the boundary conditions of the relevant transport processes occurring in the buffer, i.e. advection and diffusion.

Model studies/experimental studies

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

Natural analogues/observations in nature

If relevant, natural analogues and/or observations in nature regarding the process will be documented under this heading.

Time perspective

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

Handling in the safety assessment SR-Can

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

- neglected on the basis of the information under the previous headings,
- neglected provided that a certain condition is fulfilled, e.g. that the buffer density is within a certain range,
- included by means of modelling.

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

Time periods: On what time periods is the process relevant for the system evolution? In e.g. the case of the buffer, relevant time periods might be

- the resaturation phase extending from the time of deposition until the point in time when the buffer is fully water saturated,
- the so called thermal phase extending from the time of deposition and throughout the approximately 1,000 year time period of elevated temperature in the buffer or
- the long-term time scale extending throughout the one million year assessment time and including the varying conditions in the bedrock caused by long-term climate and other environmental variations.

By documenting the relevance of the process for applicable time periods, the process system can be simplified by omitting the process in time periods during which it is not relevant.

Boundary conditions: How are the boundary conditions handled? Are e.g. spatially and temporally varying chemical and hydraulic conditions considered?

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

The special cases of failed canister and of earthquakes altering deposition hole or tunnel geometry: These special cases imply altered conditions that could influence many processes in particular for the fuel, the canister, the buffer and the backfill and they may thus need to be discussed separately. Canister failures and earthquakes of a magnitude that could affect the deposition hole or tunnel geometry are not expected during the several thousands of years after deposition when temperate conditions are likely to prevail, meaning that the special cases are not relevant for many "early" processes. Should the assumptions regarding the lack of occurrence of significant, early canister failures or earthquakes not be corroborated by more detailed and integrated analyses in the safety assessment, this simplification of the handling of the special cases will be reconsidered.

As a result of the information under this subheading, a mapping of all processes to method of treatment and, in relevant cases, applicable models will be produced, see the Interim main report /SKB, 2004a/ for an example. The mapping will be characterised on different time scales.

Handling of uncertainties in SR-Can

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

Uncertainties in mechanistic understanding: The uncertainty in the general understanding of the process will be discussed based on the available documentation and with the aim of answering the question: Are the basic scientific mechanisms governing the process understood to a level necessary for the suggested handling? Alternative models may sometimes be used to illustrate this type of uncertainty.

Model simplification uncertainties: In most cases, the quantitative representation of a process will contain simplifications. These may result in a significant source of uncertainty in the description of the system evolution. Alternative models or alternative approaches to simplification for a particular conceptual model may sometimes be used to illustrate this type of uncertainty.

Input data and data uncertainties: The set of input data necessary to quantify the process for the suggested handling will be documented. The further treatment of important input data and input data uncertainties will, at later stages of the SR-Can project, be described in an Input data report, to which reference will be made if relevant.

References

A list of references used in the process documentation.

1.2 Documentation of participating experts and of decisions made

In this Interim process report, the experts involved in assembling the basic information about the processes are listed in Table 1-1. The sub-sections "Handling in the safety assessment SR-Can" and "Handling of uncertainties in SR-Can" have been produced by Patrik Sellin and Allan Hedin, SKB in collaboration with the expert that assembled the information on the process in question.

In the final version of the SR-Can process report, each process description will contain a documentation of which expert(s) assembled the basic information on the process, which expert(s) were involved in the decision regarding treatment in the safety assessment and the dates for the final revision of the text and for the final decision on handling. All these experts involved will be included in the forthcoming SR-Can expert database, see further the Interim main report /SKB, 2004a/.

1.3 Initial state of the buffer in SR-Can

The following is an overview description of the initial state of the buffer, i.e. its state at the time of deposition. A more formal and exhaustive account is given in the Interim initial state report of SR-Can /SKB, 2004c/. In that description, the specified set of variables describing the buffer, see section 1.4, is utilised.

In the deposition holes, the copper canister is surrounded by a buffer of clay. The buffer is deposited as bentonite blocks and rings. The blocks are placed below and above the canister and the bentonite rings surround the canister.

Two different types of bentonite are considered as reference buffer material for the purpose of SR-Can. One is a natural Na-bentonite of Wyoming type (MX-80) supplied by the American Colloid Company and the other is Calziumbentonit IBECO DEPONIT CA-N, a natural Ca-bentonite from Milos supplied by Silver and Baryte (non-converted Milos-bentonite).

The bentonite consists mainly of the smectite mineral montmorillonite (about 85%) with the characteristic property that it swells in contact with water. The montmorillonite content is not expected to vary by more than a few percent. MX-80 also contains other minerals: albite, quartz, cristobalite, muscovite and gypsum, and in addition, grains of pyrite, calcite, siderite, barite and iron hydroxides. Other minerals in Milos-bentonite are mainly calcite, dolomite, quartz, and cristobalite.

Data for the two buffer materials are summarised in Table 1-2.

There are primarily two methods available for fabrication of bentonite blocks and rings; unaxial pressing and isostatic pressing. Objects thicker than 0.5–1 m cannot be easily produced by unaxial pressing and the development of isostatic pressing is therefore important, since no equipment to fabricate full size buffer components with isostatic pressing is available in Sweden today. Fabrication of the blocks and rings by isostatic pressing requires that the objects are machined to the tolerances specified. The bentonite, bought in bulk form and transported by ship, is subject to quality control both before loading in the ship and at reception. Quality control is undertaken also during the manufacture of the blocks and rings; one important check is the water content before pressing so that this can be adjusted.

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

Process	Expert author, affiliation
Radiation attenuation/ heat generation	Allan Hedin, SKB
Heat transport	Harald Hökmark, Clay Technology
Freezing	Sven Knutsson, Luleå University of Technology
Water uptake and transport under unsaturated conditions	Lennart Börgesson, Clay Technology
Water transport under saturated conditions	Lennart Börgesson, Ola Karnland, Clay Technology
Gas transport/dissolution	Patrik Sellin, SKB
Piping/erosion	Lennart Börgesson, Clay Technology
Swelling/Mass redistribution	Lennart Börgesson, Clay Technology
Liquefaction	Lennart Börgesson, Clay Technology
Advection	Ola Karnland, Clay Technology
Diffusion	Michael Ochs, BMG Engineering
Colloid transport	Patrik Sellin, SKB
Sorption (including ion exchange)	Michael Ochs, BMG Engineering
Alterations of impurities	Jordi Bruno, David Arcos, Enviros
Aqueous speciation and reactions	Patrik Sellin, SKB
Osmosis	Ola Karnland, Clay Technology
Montmorillonite transformation	Ola Karnland, Clay Technology
Colloid release	Ola Karnland, Clay Technology
Radiation-induced transformations	Patrik Sellin, SKB
Radiolysis of pore water	Lars Werme, SKB
Microbial processes	Karsten Pedersen, University of Göteborg, Ola Karnland, Clay Technology
Speciation of radionuclides	Patrik Sellin, SKB
Transport of radionuclides in water phase	Patrik Sellin, SKB
Transport of RN in gas phase	Patrik Sellin, SKB

Table 1-2. Impurities and accessory minerals in MX-80 and Deponit CA-N bentonite.

Component	MX-80 (wt-%)	Deponit CA-N (wt-%)	Uncertainty (± wt-%)
Calcite + Siderite	0	10	1
Quartz	3	1	0.5
Cristobalite	2	1	0.5
Pyrite	0.07	0.5	0.05
Mica	4	0	1
Gypsum	0.7	1.8 (anhydrite)	0.2
Albite	3	0	1
Dolomite	0	3	1
Montmorillonite	87	81	3
Na-	72%	24%	5
Ca-	18%	46%	5
Mg-	8%	29%	5
K-	2%	2%	1
Anorthoclase	0	2	1
CEC (meq/100g)	75	70	2

The important aspect in the manufacture of bentonite blocks and rings and the subsequent deposition process is to achieve a specific final density in the water-saturated buffer. The density requirement for the saturated buffer is 1950–2050 kg/m^3. The bulk density is dependent on the slots left for technical reasons between the canister and buffer and between buffer and rock. The slot between the canister tube and the buffer is 5 mm wide and the slot along the circumferential boundary between the buffer and the rock is 30 mm. The slots are left filled with air.

The buffer emplacement in a tunnel may take place several months after the deposition holes were drilled. The deposition holes are assumed to be filled with water in the meantime, which is why draining is the first step in the preparation of the holes. Deposition starts with the hole at the far end of the tunnel. The buffer is put into position by a specially designed buffer filling vehicle. The bentonite lining is thereafter checked. The emplacement of the copper canister is done with a specially designed deposition machine which also places a top bentonite block immediately after the canister is emplaced. The emplacement of the canister will probably be documented with a photograph of the canister in its final position before the remaining bentonite blocks are emplaced. The final handling procedures and the final design of the buffer filling vehicle and the deposition machine are not decided yet, but do not affect the description of the work procedures. Small geometric tolerances in the deposition holes mean a very small risk for faulty emplacement of the buffer and canister.

The bentonite must be protected from water or high humidity until the tunnel is backfilled. The reason is that the buffer may start swelling before the deposition of the canister and before the tunnel backfilling can apply its counterweight on the buffer. One possible method is to insert a drain tube in the deposition hole and to protect the whole buffer with a plastic bag that is kept sealed until the backfilling of the tunnel starts. The plastic bag and drain tube would be removed after use.

1.4 Definition of buffer variables

Each component in the EBS initial state is described by a specified set of physical variables, selected to allow an adequate description of the long-term evolution of the component in question in the safety assessment.

The buffer is bounded on the inside by the interface towards the canister, on the outside by the interfaces towards the deposition hole, at the bottom by the interface to the bottom concrete plate and on the top by the interface towards the backfill.

The buffer as it is delimited by the variable buffer geometry is characterised thermally by its temperature and with respect to radiation by its radiation intensity, mainly γ and neutron radiation. Hydraulically, the buffer is characterised by its water content, and sometimes by gas concentrations and by hydrovariables (pressure and flows), which are mainly of interest in the phase when the buffer is being saturated with water. The buffer is mechanically characterised by its stress state.

The chemical state of the buffer is defined by the composition including the montmorillonite composition, which includes the clay minerals, and impurities. The chemical state is also defined by the pore water composition and the occurrence of structural and stray materials in the deposition hole.

The variables are defined in Table 1-3. The values of some of the variables are dependant on the density of the different phases. The following values have been used: density of water (ρ_w) is 1000 kg/m^3 and density of clay solids (ρ_{cs}) is 2780 kg/m^3.

Table 1-3. Variables for buffer.

Variable	Definition
Buffer geometry	Geometric dimensions for buffer. A description of e.g. interfaces on the inside towards the canister and on the outside towards the geosphere.
Pore geometry	Pore geometry as a function of time and space in buffer. The porosity, i.e. the fraction of the volume that is not occupied by solid material is often given.
Radiation intensity	Intensity of (α, β,) γ and neutron radiation as a function of time and space in buffer.
Temperature	Temperature as a function of time and space in buffer.
Water content	Water content as a function of time and space in buffer.
Gas content	Gas contents (including any radionuclides) as a function of time and space in buffer.
Hydrovariables (pressure and flows)	Flows and pressures of water and gas as a function of time and space in buffer.
Stress state	Stress conditions as a function of time and space in buffer.
Bentonite composition	Chemical composition of the bentonite (including any radionuclides) in time and space in buffer, levels of impurities in time and space in buffer together with mpurities also include minerals, other than smectite.
Montmorillonite composition	Mineralogical/Chemical composition of the bentonite (including any radionuclides) in time and space in buffer. Levels of impurities in time and space in buffer. Impurities also include minerals, other than montmorillonite.
Pore water composition	Mineralogical composition and structure of the montmorillonite mineral in the bentonite. This variable also includes the charge compensating cations attached to the montmorillonite surface.
Structural and stray materials	Chemical composition and quantity of concrete bottom in deposition holes.

The initial values of the variables, i.e. the values at the time of deposition, are given in the SR-Can Interim initial state report /SKB, 2004c/.

1.5 Buffer performance and safety

In order to evaluate buffer performance in the safety assessment, a number of so called function indicators and criteria that these should fulfil over time, have been formulated, see further the Interim main report, /SKB, 2004a/. The criteria are summarised in Table 1-4, where also references to the sections in this report where the criteria are motivated are given.

Table 1-4. Summary of the buffer function indicators and the criteria they should fulfil.

Function indicator	Criterion	Rationale	Reference
Bulk hydraulic conductivity	$k^{Buff} < 10^{-12}$ m/s	Avoid advective transport in buffer	2.5.2
Swelling pressure	$P_{Swell}^{Buff} > 1$ MPa	Ensure tightness, self sealing	2.5.2
Maximum temperature	$T^{Buffer} < 100°\mathrm{C}$	Ensure buffer stability	2.7.9
Minimum temperature	$T^{Buffer} > 0°\mathrm{C}$	Avoid freezing	2.4.2
Buffer density around entire canister	$\rho_{Bulk}^{Buff} > \rho_{Sink}$ kg/m^3	Avoid canister sinking (criterion to be determined)	(criterion to be determined)
Buffer density around entire canister	$\rho_{Bulk}^{Buff} > 1800$ kg/m^3	Exclude microbial activity	2.7.13
Buffer density around entire canister	$\rho_{Bulk}^{Buff} > 1650$ kg/m^3	Prevent colloid transport through buffer	2.7.4
Buffer density around entire canister	$\rho_{Bulk}^{Buff} < 2100$ kg/m^3	Ensure protection of canister against rock shear	2.6.1

1.6 Summary of handling in safety assessment

Table 1-5 summarise the handling of buffer processes in the safety assessment SR-Can, as suggested in this report. In the table, the process is either “mapped” to a model by which it will be quantified or associated with a brief verbal description of how it will be handled. Since the initial evolution, characterised by unsaturated conditions and elevated temperatures is in many respects different from the long-term, saturated phase, the description in the table has been divided accordingly.

The information in the table can be summarised as follows:

For the initial saturation phase, the peak canister and buffer temperatures and the THM evolution as the buffer saturates need to be quantified. Modelling of the thermal evolution of the entire near field will be performed with the near field system evolution model to evaluate peak canister and buffer temperatures. Coupled THM modelling of the buffer will be performed to elucidate the hydraulic evolution for different hydraulic conditions in the bedrock. The swelling at the end of the saturation phase is addressed by both the system model and the THM model, where the former can be used for rapid evaluations of the final result of the swelling for a number of input data combinations, including osmosis effects due to intruding saline water and the latter could simulate the pathway to the final result.

The chemical evolution during the thermal phase of elevated and varying temperature in the buffer will be addressed by the chemistry model.

Most other processes are neglected during the early saturation and thermal phases.

The long-term chemical evolution following the thermal phase will be addressed by both the chemistry model and the system model and for the varying boundary conditions expected as a result of climate change etc. The former model provides a spatially resolved result and involves more chemical reactions whereas the latter can be used for rapid evaluations for a number of input data combinations. Montmorillonite transformation is handled by separate modelling. Erosion due to dilute intruding groundwater during glacial conditions must be addressed if the hydrogeochemical analyses imply that ionic strengths below the given criterion cannot be excluded. Colloidal release needs to be modelled accordingly.

Table 1-5. Process table for the buffer describing how buffer processes will be handled in different time frames and in the special cases of earth quakes and failed canisters. Green fields denote processes that are neglected or irrelevant for the time period of concern. Red fields denote processes that are quantified by modelling in the safety assessment. Orange fields denote processes that are neglected subject to a specified condition.

Buffer	**Resaturation/ "thermal" period**	**Long-term after saturation and "thermal" period**	**Earthquakes**	**Notes**
Intact canister				
Radiation attenuation/heat generation	Neglected	Neglected	Neglected	
Heat transport	System model	(System model)	Irrelevant	Consider poorly centred canister?
Freezing	Neglected	Neglected if buffer temperature > 0°C. Otherwise bounding consequence calculation.	Irrelevant	Repository temperature in long term obtained from permafrost depth modelling.
Water uptake and transport for unsaturated conditions	THM model	Irrelevant by definition	Irrelevant	
Water transport for saturated conditions	Irrelevant by definition	Neglected if hydraulic conductivity < 10^{-12} m/s	Consider pressure transients	Evaluate effects on conductivity of chemical evolution and mass redistribution and of possible changes of hydraulic gradients for permafrost and glaciation
Gas transport/ dissolution	Through dissolution	(Through dissolution)	(Through dissolution)	
Piping/erosion	Model study	Irrelevant	Irrelevant	
Swelling/Mass redistribution	THM modelling including interaction buffer/backfill and thermal expansion System model (final swelling)	Integrated evaluation of erosion, convergence, corrosion products, creep, swelling pressure changes due to ion exchange and salinity, canister sinking	Part of integrated assessment of buffer/canister/rock	Need to also consider deviations in amount of buffer initially deposited.
Liquefaction	Irrelevant	Irrelevant	To be determined	
Advection	Simplified assumptions of mass transport of dissolved species during saturation.	Neglected if hydraulic conductivity < 10^{-12} m/s	Consider pressure transients	See "Water transport for saturated conditions"
Diffusion	Chemistry model (thermal, saturated phase; unsaturated phase disregarded)	Chemistry model System model	Consider altered geometry (diffusion pathways).	Consider varying ground water compositions Thermal transient in chemical model?
Sorption (including ion-exchange)	Chemistry model (thermal, saturated phase; unsaturated phase disregarded)	Chemistry model System model		
Alterations of impurities	Chemistry model (thermal, saturated phase; unsaturated phase disregarded)	Chemistry model System model		

Pore water speciation	Chemistry model (thermal, saturated phase; unsaturated phase disregarded)	Chemistry model System model	
Osmosis	System model (initial swelling)	Evaluation through comparison with empirical data System model	Handling of long-term intrusion of saline water
Montmorillonite transformation	Model calculations (thermal, saturated phase; unsaturated phase disregarded)	Model calculations	
Colloid release/erosion	Neglected	Neglected if $[M^{2+}]$ > 1 mM Otherwise modelled	
Radiation-induced transformations	Neglected	Neglected	
Radiolysis of pore water	Neglected	Neglected	
Microbial processes	Neglected if ρ > 1800 kg/m^3, otherwise quantitative estimate of sulphate reduction	Neglected if density > 1800 kg/m^3, otherwise quantitative estimate of sulphate reduction	
Failed canister			
Gas transport/ dissolution	Irrelevant	Quantitative estimate based on empirical data	
Colloid transport	Irrelevant	Neglected if density < 1650 kg/m^3, otherwise bounding calculation	
Speciation of radionuclides	Irrelevant	Assumptions based on empirical data	
Transport of radionuclides in water phase	Irrelevant	COMP23 Analytic	
Transport of radionuclides in gas phase	Irrelevant	Quantitative estimate based on empirical data	

The effects of the chemical evolution on key properties like swelling pressure and hydraulic conductivity will be evaluated using empirical relationships. A number of issues related to mass redistribution in the buffer need to be evaluated for the long-term evolution.

In the case of a canister failure, the release of gas from the corroding cast iron canister insert will be handled by quantitative estimates based on experimental studies of gas transport through bentonite. Diffusion of radionuclides through the buffer is calculated with radionuclide speciations, necessary for the selection of diffusion and sorption data, estimated on the basis of experimental data. Transport of fuel colloids through the bentonite is neglected if the buffer density exceeds a specified value, otherwise the effect of this process on dose consequences is estimated by a bounding calculation case.

2 Buffer processes

2.1 Radiation-related processes

2.1.1 Radiation attenuation/heat generation

Overview/general description

γ and neutron radiation from the canister are attenuated in the buffer. The radiation that is not attenuated reaches out into the near-field rock.

The radiation is of importance for the chemical processes radiation-induced montmorillonite decomposition and γ radiolysis of pore water.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	Yes	Yes	
Temperature	No	Negligible compared to other processes influencing temperature	
Water content	Yes	No	
Gas content	Negligible	No	
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	Yes	No	
Pore geometry	No	No	
Swelling pressure	No	No	
Smectite composition	Yes		
Pore water composition	Negligible		
Smectite content	Yes	Negligible	
Impurity content	No	Negligible	
Structural and stray materials	No	Negligible	

The magnitude of the attenuation is dependent above all on the geometry, density and water content of the buffer.

The result is a radiation field in the buffer that can lead to radiolysis and that has a marginal impact on the montmorillonite, see further section 2.7.11.

Attenuation of γ and neutron radiation will raise the temperature of the buffer, but the effect is negligible compared with other temperature-raising processes.

Boundary conditions

The boundary condition is the radiation intensity to which the buffer is exposed, i.e. the flux leaving the canister outer surface.

Model studies/experimental studies

Attenuation of γ and neutron radiation can be calculated theoretically for an arbitrary material if geometry and composition are known, see e.g. the model studies mentioned for the corresponding process in the canister. Experimental studies of radiation attenuation in bentonite are reported in section 2.7.11.

Natural analogues/observations in nature

Not applicable.

Time perspective

The time perspective is determined by the decay properties of the spent fuel, to be described in the process report for the fuel, see the SR 97 version of the process report /SKB, 1999/.

Handling in the safety assessment SR-Can

Quantitatification of this process is needed to evaluate the processes 2.7.11 Radiation-induced transformations and 2.7.12 Radiolysis of pore water. The former is shown to be negligible in section 2.7.11, for the relevant radiation intensities. The latter could affect the canister surface, but also this effect is shown to be insignificant, see further sections 2.7.12 and the relevant canister process /SKB, 1999/.

Time periods: The γ and neutron radiation is significant during approximately 1,000 years.

Boundary conditions: Any quantitative treatment will have to include the radiation flux leaving the canister outer surface as a boundary condition.

Influences and couplings to other processes: The geometry and the material compositions influence this process. Simple bounds can be put on the effects of temporal variations of the material compositions.

The special cases of failed canister and of earthquake: Canister failures and earthquakes of a magnitude that could affect the process are not expected during the roughly 1,000 year time period in which there is any significant radiation. The process is thus not handled for these cases. Should the assumptions regarding significant, early canister failures or earth quakes not be confirmed by the results of the safety assessment, the handling of the special cases will be reconsidered.

Uncertainties

Uncertainties in mechanistic understanding

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

Model simplification uncertainties for the above handling in SR-Can

Not relevant.

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

In general, data for a quantitative description of the process are known with sufficient accuracy for the needs of the safety assessment, considering the small effects of this process on the repository system.

2.2 Thermal processes

2.2.1 Heat transport

Overview/general description

Heat is transported from the canister surface to the buffer, through the buffer and finally from the buffer to the rock both directly and via the tunnel backfill. The first time after deposition there will be gaps between the canister and the buffer and between the buffer and the rock. During this time, heat will be transported by a combination of radiation and conduction across the two gaps and by pure conduction across the bentonite which will be incompletely saturated. There may be some convection, but because of the low permeability of the bentonite and because of the narrow gap geometries, this contribution can be ignored. In particular the heat resistance of the gap between canister and bentonite will be high because of the low emissivity of the copper surface and the associated low radiant heat transfer. The heat resistance of the gap between buffer and rock will be lower and will not have such an impact on the canister temperature as the canister/buffer gap. I addition, the buffer/rock gap will start to close soon after deposition while the canister/buffer gap will remain open longer. The efficiency of the heat transport through the buffer region is important for the performance of the system, since it affects two important temperatures: that of the canister surface and that in the buffer itself.

When the buffer has been water-saturated and has swelled out so that all gaps and joints are filled, all heat transport takes place by conduction though water-saturated bentonite.

A few days after deposition, heat transport through the buffer is largely independent of the buffer heat capacity, and heat conduction in the buffer can be approximately described by the time-independent heat conduction equation:

$$\nabla(\lambda\nabla T) = 0$$

The most important parameter is thus the thermal conductivity, λ, of different parts of the system. The thermal conductivity of bentonite is primarily dependent on density, water saturation and mineral composition. The thermal conductivity of water-saturated MX-80 bentonite that has swelled out to its intended density, 2,000 kg/m^3, is about 1.2 W/(m·K) according to laboratory experiments.

The thermal conductivity of gaps filled with water is 0.60 W/(m·K), while that of gaps filled with gas, i.e. air and vapour, is about 0.03 W/(m·K). For the canister/bentonite gap the effective conductivity is 0.03–0.06 W/(m·K) taking into account also the contribution from the radiant heat transfer /Hökmark and Fälth, 2003/.

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

Variable	Process Influencing?	Process Influenced?	Notes
Radiation intensity	No	No	
Temperature	Yes	No	
Water content	Yes	Yes	
Gas content	No	No	
Hydro variables (pressure and flows)	Yes	No	
Buffer geometry	No	Yes	
Pore geometry	Yes	Yes	
Stress state	No	No	
Bentonite composition	No	Yes	May affect thermal conductivity
Montmorillonite composition	No	No	
Pore water composition	No	No	
Structural and stray materials	No	Yes	May affect thermal conductivity

Temperature: The influence of heat transport on the buffer temperature is obvious. If the heat transport is efficient, the temperature in the inner parts of the system will be lower than if there is a high thermal resistance. The temperature in the outer parts, however, is practically independent of the conditions in the interior of the deposition holes. Different models have been proposed for estimating the influence of the temperature on the thermal conductivity of bentonite /Knutsson, 1983/. The effect is a small but uncertain increase, which can be pessimistically ignored.

Water content: The conductivity of bentonite with a high montmorillonite content can be estimated with the aid of the Equations (1) to (3), which give the thermal conductivity as a function of the degree of saturation Sr and the porosity n, which, together with the mineral density, determine the density of the buffer /Knutsson, 1983/.

$$\lambda = \lambda_0 + K_e (\lambda_1 - \lambda_0) \qquad (1)$$

where

λ_0 = thermal conductivity at $S_r = 0$ (dryness)

λ_1 = thermal conductivity at $S_r = 100$ percent (water saturation)

$K_e = 1 + \log S_r$

λ_0 and λ_1 are determined according to Equations (2) and (3):

$$\lambda_0 = 0.034 \cdot n^{-2.1} \qquad (2)$$

$$\lambda_1 = 0.56^n \cdot 2^{(1-n)} \qquad (3)$$

Equation (1) gives values with an estimated accuracy of 20 percent and is valid for porosities larger than 0.15. A number of similar expressions relating thermal conductivity to saturation and porosity can be found in the literature /Börgesson et al, 1994/.

The thermal gradients in the buffer will influence the state of saturation, such that the water content of the innermost parts of the buffer will decrease temporarily.

Gas content: The heat transport will not affect the gas content. The gas content in itself does not have any influence on the heat transport. If there is a gas phase, the effects of this will be just those of the incomplete saturation.

Hydro variables: Temperature variations will cause changes to the pore water pressure.

Buffer geometry: The resistance to heat transport in the buffer is dependent on the distance between the canister and the deposition hole boundary, which entails changes in the temperature of the buffer if the dimensions of the canister hole or the location of the canister in the hole should be altered. After water saturation, however, the variations in geometry that could occur due to block fallout or a poorly centred canister have negligible effects. This is also true of effects of instantaneous or time-dependent deformations that can alter the geometry of the canister hole. Before water saturation, a poorly centred canister can be of importance for the distribution of gas-filled volumes and therefore for the temperature level and temperature distribution in the buffer.

Pore geometry: The geometry of the pore system will change slightly because of thermal expansion. The porosity, i.e. the volume fraction taken up by pores, influences the thermal conductivity. Equations (1) to (3) give approximate relations between porosity and conductivity. The porosity of water-saturated highly-compacted MX-80 bentonite with a density of 2,000 kg/m^3 at water saturation is 0.44, and the thermal conductivity according to Equation (3) is 1.14 W/(m·K).

Swelling pressure: Different models have been proposed for estimating the influence of increasing pressure on the conductivity of bentonite /Knutsson, 1983/. They show a slight increase of the thermal conductivity, but the effect is not sufficiently verified to be credited /Börgesson et al, 1994/.

Smectite composition: There is no dependence on the composition of the smectite.

Smectite content: The smectite content per se is not decisive for the buffer's thermal conductivity, since the most common accessory minerals have a similar thermal conductivity. An exception is quartz, which has a higher thermal conductivity.

Impurity content: The presence of impurities may affect the thermal conductivity. To be of any importance, the impurity content must be high and the thermal properties of the impurities significantly different from those of the bentonite.

Structural and stray materials: To be of any importance to the heat transport, there must be very significant amounts of non-buffer materials.

Boundary conditions

There are two boundary conditions:

- The heat flux from the local canister, determined by the power at the time of deposition and by the heat decay characteristics of the waste,
- The temperature at the walls of the deposition hole.

The rock wall temperature depends not only on the local canister but also on the contribution from other canisters, on the rock thermal properties and on the repository layout.

During the saturation phase, the mode of heat transfer across the canister/buffer interface will be uncertain and complex. Also at the outer interface, between buffer and deposition hole wall, gaps will occur during the saturation phase. After saturation, there will be direct thermal contact between canister and buffer and between buffer and rock, and all heat transfer will take place by pure conduction.

Heat transport in transition canister/buffer: If the gap between the buffer and the surface of the canister is water-filled, transport across the gap takes place by conduction with a thermal conductivity of about 0.6 W/(m·K), which differs insignificantly from the thermal conductivity of partially unsaturated bentonite. If the gap is gas-filled, the equivalent thermal conductivity is between 0.03 W/(m·K) and 0.06 (m·K) and a considerable temperature differential can arise between canister surface and buffer. The size of the differential depends on how efficiently heat can be transferred by radiation, i.e. on the emissivity of the canister surface, and on how well the canister is cooled via other surfaces where there are no gas-filled gaps. The size of the temperature differential can be up to 17°C if the gap is 10 mm wide /Hökmark and Fälth, 2003/.

Heat transport in transition buffer/rock: The space between buffer and rock will be affected by water uptake and swelling significantly earlier than the space between canister and buffer. The radiant heat flux across open buffer/rock gaps will be more efficient because of the high emissivities of the two opposing surfaces. Results from the SKB prototype repository indicate that the temperature offset may be about 5 or 6 degrees a few months after deposition for a 50 mm gap that is filled with bentonite pellets initially /Goudarzi and Johannesson L-E, 2004/.

Model studies/experimental studies

Model studies. Predictions of the temperature evolution in the near field have been done analytically /Claesson and Probert, 1996; Hökmark and Fälth, 2003/ and numerically /Thunvik and Braester, 1991; Hökmark, 1996; Ageskog and Jansson, 1999/.

The temperature at the buffer/canister transition reaches a peak after about 10–30 years, depending on layout and rock thermal properties. A peak is reached at the buffer/rock transition after about 30–50 years.

The temperature levels that will apply at the deposition hole wall are dependent on the thermal diffusivity of the rock, the canister power, the tunnel spacing and the canister spacing. By means of analytical solutions it is possible to calculate the temperature in the rock at different points around a canister for arbitrary repository geometries /Claesson and Probert, 1996; Hökmark and Fälth, 2003/. For example: if the canister spacing is 6 m, the tunnel spacing 40 m, the canister power at the time of deposition 1700 W and the rock thermal conductivity 2.8 W/ (m·K), the maximum temperature increase at the wall of the deposition hole will be 50°C.

The temperature levels that will apply in different parts of the buffer and in the canister/buffer transition are further dependent on the heat conduction properties of the buffer and on the possible presence of gaps and drying cracks. If it is assumed that the effective thermal conductivity between canister and rock is 0.7 W/(m·K), the temperature difference between them will be 30°C at an initial canister power of about 1700 W /Thunvik and Braester, 1991/. The difference decreases with time and is about 10°C after 100 years and only about a degree after 1,000 years.

The duration of the temperature pulse is a few thousand years. After 2,000 years the excess temperature in the repository is about 20°C and after 5,000 years about 15°C /Hökmark, 1996/. No gradients exist in the near field after about 2,000 years.

Experimental studies: /Börgesson et al, 1994/ measured the thermal conductivity of samples of compacted MX-80 bentonite by use of a heat pulse technique, Figure 2-1. The results verified the dependence on saturation and porosity suggested by the general expressions (Equations 1 through 3). The results accord well with corresponding results obtained for similar materials, for instance the bentonite tested in the FEBEX experiment /Huertas et al, 2000/.

The Prototype Repository includes six full-scale deposition holes in two tunnel sections separated by a concrete plug /Goudarzi and Börgesson, 2003/. There are two deposition holes in the outer section and four in the inner one. Inflow measurements performed prior to bentonite emplacement and start of the actual test showed that there are very significant differences between the individual holes. The supply of water to the two holes in the outer section is sufficiently low that the bentonite buffer is not close to saturation about 6 months after test start. The temperature drop across the still open canister/bentonite space in the dry holes is about 19°C. In at least one of the holes in the inner section, there are indications that that the annular space between canister and rock is almost saturated about two years after test start.

The effective thermal conductivity λ_{eff} of the buffer in the annular space between canister and rock wall at canister mid-height can be estimated by use of the following expression:

$$\lambda_{eff} = \frac{q_c}{\Delta T} \cdot R_c \cdot \ln \frac{R_2}{R_1}$$

Here ΔT is the temperature difference between two points at distances R_1 and R_2, respectively, from the heater axis. R_c is the canister radius. The canister surface heat flux q_c at canister mid-height must be estimated from the total power and corrected to account for the non-uniform distribution of the heat output over the canister surface /Hökmark and Fälth, 2003/. Applying the expression to the three holes described above using data reported by /Goudarzi and Börgesson, 2003/ gives bentonite conductivity values ranging between 1.1 for the inner hole and 1.2 W/(m·K) for the two outer holes.

A similar estimate of the thermal conductivity of the bentonite in the canister retrieval test /Goudarzi et al, 2003/ gives a value of about 1.1 W/(m·K) 300 days–800 days after test start.

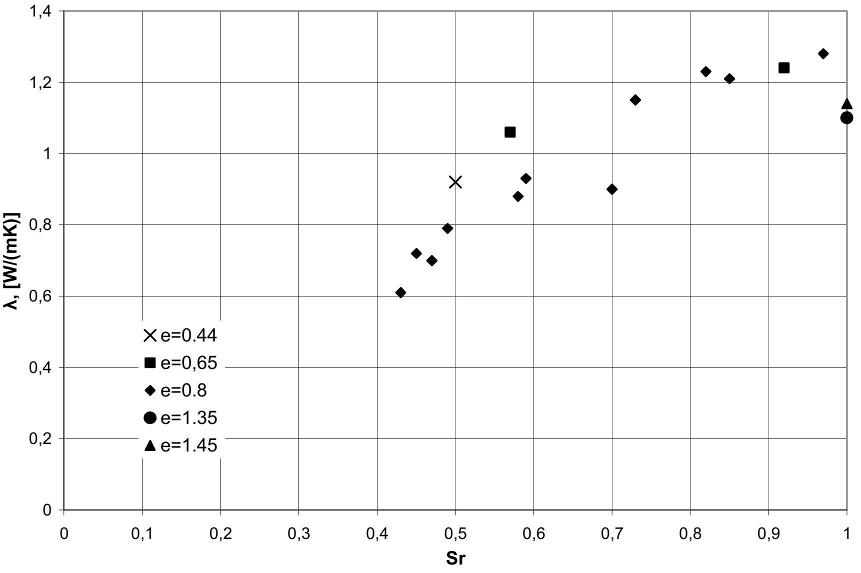


***Figure 2-1.** Heat conductivity of laboratory–scale bentonite samples as function of saturation for a few values of the void ratio e.*

With respect to order of magnitude, the results of the field tests verify the results of the laboratory tests. However, the difference between results for the bentonite at about 80% saturation in the outer section of the prototype repository and the bentonite at almost 100% saturation in the almost saturated hole in the inner section and in the canister retrieval test is not consistent with the theoretically and experimentally derived saturation-conductivity relations. Variations caused by uncertainties in power, power distribution and instrument positions overshadow effects of differences in saturation and point to the influence of gaps and inhomogeneities on the organization of heat flow around the canisters.

Natural analogues/observations in nature

Not applicable.

Time perspective

There are two relevant time perspectives: the water saturation perspective and the heat production perspective. The time it takes to achieve full water saturation is dependent on, inter alia, the pressure conditions in the groundwater in the near field. The process is estimated to take a number of years, see section 2.5.1. After that, all heat transport takes place by conduction and under well-defined conditions with known thermal conductivities. Before then, heat transport can be influenced by the presence of gaps and joints.

After a few thousand years the heat production, and thereby the heat transport through the buffer, will have been reduced to a few percent of their original values.

Handling in the safety assessment SR-Can

The integrated thermal evolution of the fuel, cast iron insert, copper canister, buffer and rock will be modelled with the SR-Can system evolution model /Hedin, 2004/.

An important purpose of the modelling is to evaluate the peak canister surface temperature that must not exceed 100°C, taking all relevant uncertainties into consideration. These include the possible presence of gaps between canister and buffer and between buffer and rock and data for describing heat conduction over these. Critical uncertain data for this evaluation will be given in the SR-Can data report. Another purpose is to obtain an estimate of the peak buffer temperature and of the buffer temperature as a function of time.

Time periods: The modelling will encompass at least 10,000 years, but the critical time scale is that during which the peak canister temperature is reached, i.e. the first tens of years. For time scales beyond a few thousand years, it is important to determine if permafrost conditions could lead to freezing of the buffer, see further section 2.4.2. This is done by a comparison of the rock thermal evolution during permafrost conditions to the buffer freezing temperature, taking the residual power of the canisters into account if relevant.

Boundary conditions: The treatment of heat transfer over the buffer boundaries is described in /Hedin, 2004/ and /Hökmark and Fälth, 2003/. As a special case, a poorly centred canister will be considered.

Influences and couplings: The process is coupled to the water saturation process which is difficult to model in detail, especially the evolution of gaps. Also, the water saturation process is highly dependent on the uncertain hydraulic conditions in the rock around the deposition hole. The peak canister temperature will therefore be determined under the assumption that no additional water is taken up by the buffer after deposition, i.e. a time independent, pessimistically chosen heat conductivity is used. Hereby, the influences of varying hydraulic conditions in the saturating buffer, including porosity variations, are pessimistically neglected. The thermal conductivity of the buffer will also be selected considering the possible influence of impurity contents and presence of stray materials in the system. The gap between the canister and the buffer will pessimistically be assumed to be open until the peak temperature has been reached.

The special cases of failed canister and of earthquakes: Canister failures and earthquakes of a magnitude that could affect the thermal evolution are not expected during the roughly 1,000 year time period in which the buffer temperature evolves significantly. The process is thus not handled for these cases. Should the assumptions regarding significant, early canister failures or earth quakes not be confirmed by the results of the safety assessment, the handling of the special cases will be reconsidered.

Uncertainties

Uncertainties in mechanistic understanding

Heat transfer from the hot canisters via the buffer to the near-field rock and backfill is in principle a simple process that can be described with reference to basic laws of physics. The character of the subprocesses that participate in the heat transport is also known and the theoretical modelling of their function is based on well-known physical principles.

Model simplification uncertainties for the above handling in SR-Can

Several pessimistic modelling simplifications regarding the coupling to the hydraulic evolution are described under "Handling in the safety assessment" above. The pessimistic simplifications are justified since the prime purpose of the modelling is to obtain upper limits on canister and buffer temperatures. Sensitivity analyses will in some cases shed light on the effects of the simplifications.

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

Critical input data for the modelling of this process will be given in the SR-Can data report. Critical uncertain input data in determining the peak canister temperature include the heat power of the fuel, the thermal conductivity of the partly saturated buffer material and of the host rock. Also uncertainties of the emissivities of the inner buffer and in particular the outer copper surfaces are decisive in determining the peak canister temperature. Sensitivity calculations will be done to elucidate the influence of various data uncertainties.

2.2.2 Freezing

Overview/general description

When water freezes, energy is released (333 J/g) and volume expansion takes place (approximately 9%). In a porous medium containing water in the pores, water will freeze if the temperature is below the freezing point. However, if the geological material contains enough fines, there will also be an inflow of additional water to the frost front. This is caused by the thermo-dynamic situation close to the silt and clay particles. When this additional water enters regions with freezing temperatures, the water will freeze. Thus more water is built into the material, during the freezing process, than can be stored in the existing pore system. This additional water, which also turns into ice and expands, will thus give rise to a volume expansion of the material. This process is known as frost heaving or segregation freezing, in contrast to the freezing of the water in the existing pores (in-situ freezing).

Bentonite consists, to a large extent, of highly active clay minerals with a large specific surface. Geological material consisting of this type of clay minerals fulfill the basic requirements for being frost active and show consolidation effects when thawing. However, ice lensing and thus frost heave, can only take place if water is available and the hydraulic conductivity is high enough to let the water through to the frost front and the frozen parts of the system. If the time available is sufficiently long, frost heave and thus the corresponding volume expansion, will take place even though the hydraulic conductivity is extremely small. During a normal winter in a soil volume close to the ground surface, there will not be enough time to let water through a material with a very low hydraulic conductivity and thus frost heave will normally not take place. Therefore, clays are normally classified as low to medium frost susceptible materials. But, if there is enough time, clay will exhibit great frost heave. The more fine grained the soil, the greater frost heave, but this is only valid if water is available and the time for the water transportation is sufficiently long. The time necessary for the process will depend on the hydraulic conductivity.

If the freezing soil cannot expand freely, then a pressure, sufficiently high for preventing the water from freezing, is built up. This pressure is in the range of several MPa/°C.

The freezing point is difficult to determine experimentally for the relevant conditions. An upper bound on the freezing point is however the freezing point of water i.e. approximately 0°C.

If frost heave takes place in a granular material, relocation of particles normally also occurs. Naturally occurring phenomena include sorting of particles, lifting of stones and boulders, relocation of different embedded objects etc. In a “normal” winter situation, where a geological material freezes, stones etc are lifted and relocated to about 5–10 mm a year. If the freezing goes on for thousands of years, the relocation can be significant, even though the frost action is fairly limited.

When thawing occurs, the ice melts and water is drained off. During frost heaving, ice lenses have been formed. Thus, fractures and openings have been created in the soil matrix. These fractures and openings *might* be closed during thaw. However, there are numerous examples of soil/bentonite mixtures where the hydraulic conductivity have been increased after freezing and thawing, thus indicating no closing of the fractures. Other examples show no effect of the freezing process on the hydraulic conductivity.

Suction is created in a freezing soil, causing an increased effective stress in the material and a resulting consolidation. Such a temperature induced consolidation of the material causes a volume reduction during thaw and thus fractures and openings are generated. This process takes place in soft, clayey soils. Whether there is any risk for such a process in the highly compacted bentonite is not known. It is not likely to occur due to the high density of the compacted bentonite, but in a mixture of bentonite and crushed rock it is on the other hand likely to occur.

During thaw, water is drained off if there has been a frost heaving process with ice lens formation during the freezing period, provided that water can escape. If this is not the case, water will stay in the system and thus create a looser material than expected. The buffer materials will in such a situation have a highly reduced strength and a completely different stress-strain characteristic, compared to the original material.

In summary, potential effects in buffer materials due to freezing and thawing include

- Increased stress levels during freezing.
- Frost heave if water and long time periods are available.
- Risk of relocation of embedded objects.
- Creation of fractures and openings, which *might* be closed after thaw.
- Changes in hydraulic conductivity after freezing and thawing; at least in the sand/bentonite mixture.
- Temperature induced consolidation effects in a mixture of bentonite and crushed rock.
- Different stress/strain relation after freezing and thawing.

Handling in the safety assessment SR-Can (Preliminary)

The process is neglected if the near-field of the repository has a temperature which is higher than the freezing point of bentonite, i.e. 0°C.

If this is not the case, canister integrity cannot be guaranteed, based on the above, preliminary, evaluation. It is noted that this would apply to all canisters in the repository simultaneously. The consequences of this case can be estimated by bounding calculations.

2.3 Hydraulic processes

2.3.1 Water uptake and transport under unsaturated conditions

Overview/general description

Water transport in the buffer under unsaturated conditions is a complex process that is dependent on, inter alia, temperature, smectite content, degree of water saturation and water content in the different parts of the buffer. The most important driving force for water saturation under deep repository conditions is a negative capillary pressure in the pores of the buffer that leads to water uptake from the surrounding rock. The supply of water in the rock is also a decisive factor for the temporal evolution of the process.

Following is a detailed description of the process. The degree of detail is appropriate to indicate a general understanding, but is not necessary for the needs of the safety assessment to, for example, model the main features of the hydraulic evolution of the buffer.

The following hydraulic sub-processes can be distinguished:

- Transport of water in liquid phase, which is controlled and driven by

  A1) a pressure gradient in the water,

  A3) an osmotic gradient,

  A4) gravity (coupled to A1),

  A5) a density gradient (coupled to A1).

- Transport of water in vapour form, where transport is controlled and driven by
  B1) a pressure gradient in the water,
  B2) a temperature gradient,
  B3) an osmotic gradient,
  B4) gravity,
  B5) a density gradient.
- Phase transitions between water and vapour by:
  C1) evaporation,
  C2) condensation.
- Thermal expansion of
  D1) water,
  D2) air.
- Compression of
  E1) water,
  E2) air.
- Transport in water of
  F1) dissolved air.

These processes are coupled, and are in some cases different sides of the same process. They can be described and modelled in different ways. An important variable is the pore water pressure, which can be described according to

$$\psi=(u_a-u_w)+\pi$$

where ψ is the free energy of the soil water (usually named total suction), u_w is the pore water pressure, u_a is the pore gas pressure and π is the osmotic suction. (u_a-u_w) is called the matric suction.

The total suction can thus be described from a geotechnical viewpoint as the sum of the matrix suction, which can be said in simplified terms to stem from capillary effects, and the osmosis suction, which is dependent on differences in ionic concentration.

The driving force for water flow is usually considered to be the pore water pressure, which means that the matric suction and the pore gas pressure are important variables in the modelling of water flow in water unsaturated materials. The total suction is a function of the water content of the clay (called retention curve).

The understanding of the processes that drive water transport in unsaturated buffer and backfill materials and the knowledge of how the processes are influenced by different factors are not comprehensive. The following description pertains chiefly to the models that are used for non-swelling soil materials /Fredlund and Rahardjo, 1993/. It has not yet been fully clarified whether the approach is sufficiently all-inclusive to fully describe the processes in swelling clay minerals. However, the understanding is sufficiently good to carry out reliable model calculations in the safety assessment.

A1. Transport of water in liquid phase, which is driven by a water pressure gradient, can be described by a refined version of Darcy's law. The parameters that control the process are the material's hydraulic conductivity K and the difference in pore water pressure u_w. Both of these can be described as functions of the void ratio e, the degree of saturation S_r, the ion concentration in the pore water i_c and the temperature T. The pore water pressure of unsaturated clays is also a function of the wetting history, i.e. whether the material undergoes wetting or drying, and the total average external stress.

The driving force for the liquid-phase water flow is the pore water pressure gradient. The hydraulic conductivity varies strongly with the degree of saturation.

A3. Transport of water in liquid phase, which is driven by an ion concentration gradient, osmosis. Significant osmotic effects can arise at high salinities in the groundwater and in conjunction with salt enrichment in the buffer. The driving force should be visualised as a diffusion process driven by an osmotic gradient.

A4. Transport of water in liquid phase, which is driven by gravity. The influence of gravity is reflected in the hydraulic head and thus taken care of by process A1. Gravimetrically generated flow is probably negligible in both the buffer and backfill materials due to the low hydraulic conductivity, unless the liquid pressure originates from the water pressure in the rock.

A5. Transport of water in liquid phase, which is driven by a density gradient in the water. Differences in the density of the water can be caused by e.g. differences in temperature or salinity. Density-generated flow is probably negligible in buffer material, cf. process A1.

B1. Transport of water in vapour phase, which is driven by a vapour pressure gradient. The vapour pressure u_v in the fraction of the pores that are air-filled or the relative humidity ($RH=u_v/u_{vo}$ where u_{vo} is the saturation pressure in the air) is in equilibrium with the total suction potential in the liquid phase. If RH is higher than the equilibrium relationship, water vapour condenses and increases the quantity of liquid, which in turn reduces the total suction until equilibrium arises. If the total suction differs in different parts of a soil, for example due to different water ratios, this leads to different vapour pressures, and transport of water vapour in the unfilled portion of the pores takes place with the vapour pressure difference as a driving force. At similar temperature and other circumstances water vapour is thus transported in the gradient of the relative humidity and (due to the direct correlation) in the gradient of total suction. However, this transport is parallel to process A1, which takes place in liquid phase. The relationship between these transport mechanisms has not been fully investigated.

B2. Transport of water in vapour phase, which is driven by a temperature gradient, appears to be the most important process for vapour transport. This process can also be regarded as a part of process B1. A temperature difference brings about a vapour pressure difference which transports vapour from a warmer part to a colder part. It can also be modelled as a diffusion process driven by a temperature difference /Philip and de Vries, 1957/.

The vapour transport causes a water ratio difference and thereby a difference in suction potential, which in turn leads to a liquid-phase transport in the opposite direction. In a closed system, with a temperature gradient and constant mass of water, an equilibrium situation arises after a time where the vapour transport is equal to the liquid transport in all parts. A steady state arises with water ratio differences that reflect the temperature differences in the system.

B3. Transport of water in vapour phase, which is driven by an osmotic gradient. This is an indirect process since the vapour is driven by a difference in vapour pressure that can be settled by a difference in osmotic pressure.

B4 and B5.

Transport of water in vapour phase, which is driven by gravity or a density gradient. These processes are presumably insignificant in the buffer's pore system, but may be of great importance in gaps, particularly in the vertical gap between canister and bentonite.

C1 and C2.

Phase transitions via evaporation and condensation are constantly occurring in a temperature gradient where evaporation in the warmer part is accompanied by condensation in the colder part. Aside from the fact that it can lead to heat transport, it is only significant as evaporation if the buffer is not isolated from the atmosphere. It can occur if a very dry deposition hole is allowed to stand open towards the tunnel for a very long time or if the backfill is dry.

D1 and D2.

Thermal expansion of water in liquid and vapour phase can indirectly affect the flow by changing either the water pressure or the density (convection). Other consequences are dealt with under mechanical processes.

E1. Compression of water in liquid phase is of little importance for the liquid transport processes other than for a water-saturated system, where the size of the pressure build-up associated with a temperature increase is dependent on the compressibility of the water.

E2. Compression of air can be an important factor in the water saturation process (Boyle's law). If the air is trapped, it must be compressed and go into solution in the water in order for water saturation to be achieved. In highly compacted bentonite, the suction potential appears to be sufficiently high for this to occur and full water saturation is achieved without any external water pressure. Highly-compacted pure bentonite is thus able to achieve water saturation by itself. The backfill, however, has such low suction that a pressurized air volume will remain if the air cannot seep out. A large part will disappears when full water pressure has developed.

F1. Transport of dissolved air in water. When trapped air is compressed during the water saturation phase, a portion of the volume is dissolved in the water. This solubility (Henry's law) is approximately 2 percent at air temperature but decreases rapidly at increasing temperature. In order for additional air to be dissolved, the dissolved air must be transported away, which takes place by diffusion.

Influencing/Influenced variables

Influence of salinity on wetting rate: High salinity in the groundwater that is absorbed from the near-field rock limits the swelling of the bentonite grains and initially leaves wider passages for penetration of water. The hydraulic conductivity increases with increasing salinity especially for low densities. An advantage is that saturation of the buffer can take place faster than if low-salinity water is absorbed.

The salinity of the groundwater influences the vapour pressure relation and thereby the water saturation process. For typical Swedish groundwater conditions, the effect in the buffer is small, but the influence of saline groundwater may be of decisive importance for water uptake in the backfill.

Influence of temperature on wetting rate: The viscosity of water is affected by the temperature, which means that the hydraulic conductivity increases with increasing temperature. This means that an increased overall temperature in the buffer increases the wetting rate.

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	Yes	Yes	Influencing through the water content
Water content	Yes	Yes	
Gas content	Yes	Yes	
Hydro variables (pressure and flows)	Yes	Yes	
Buffer geometry	Yes	Yes	Influencing through swelling of the blocks
Pore geometry	Yes	Yes	
Stress state	Yes	Yes	
Montmorillonite composition	No	Yes	Influenced through the retention curve
Pore water composition	Yes	Yes	
Bentonite composition	No (unless via mineral conversion)	Yes	Influenced through the retention curve
Structural and stray materials	No (unless via chemical conversion)	Yes (insignificant)	Influenced through the retention curve

Influence of water and gas content: The hydraulic conductivity is strongly affected by the degree of saturation. A commonly used relation between hydraulic conductivity and the degree of water saturation is

$$K_p = S_r{}^{\delta} K$$

where

K_p = hydraulic conductivity of partly saturated soil (m/s)

K = hydraulic conductivity of completely saturated soil (m/s)

δ = parameter ≈ 3 for MX-80 at buffer densities

The vapour diffusivity is also affected by the degree of saturation.

The pore water pressure in an unsaturated buffer is strongly affected by the water content (retention curve). For example: at $w = 10\%$ the pore water pressure is $u \approx -70$ MPa while at $w = 20\%$ the pore water pressure is $u \approx -20$ MPa. Under volume constrain the negative pore water pressure is reduced during water uptake and goes to 0 at complete water saturation.

Boundary conditions

Interaction with the rock: A key issue for the saturation process is the interaction between rock and buffer. If water is only conducted to the buffer in the water-bearing fractures and the rest of the water has to go through the rock matrix, water saturation can be both uneven and take a long time. If there is a permeable excavation-disturbed zone (EDZ) at the wall of the depoisition hole, which distributes water from the fractures along the rock wall, the wetting will be faster and more even. However, performed tests indicate that there is a small disturbance of the rock caused by the deposition hole boring, which reaches a few cm into the rock but also that the disturbance is not of significant importance for the wetting. See e.g. /Autio et al, 2003/.

Interaction with the backfill: If the backfill is composed of a mixture of bentonite and crushed rock or similar, the suction is much higher in the buffer material than in the backfill. This means that water will be transported from the backfill to the buffer and thus wet the buffer and dry the backfill. If the wetting of the backfill from the rock is faster than the wetting of the buffer, this water will also be redistributed to the buffer and thus help the wetting of the buffer.

Model studies/experimental studies

A preliminary material model containing unsaturated water flow has been devised /Börgesson and Johannesson, 1995/ and is being developed. The calculations are performed using the finite element method with the program ABAQUS. Certain general parts of the model are described in the manual /Hibbit et al/, while other parts are tailor-made for SKB. See e.g. /Börgesson and Johannesson, 1995; Börgesson and Hernelind, 1999; Börgesson et al, 2004/. The model includes the processes A1, A4, A5, B2, D1 and E1, which are the most important ones for the buffer. It is a completely coupled thermo-hydro-mechanical model and thus also includes thermal and mechanical processes in the buffer. It does not, however, include the processes that handle air (except for B2), which can be important for the saturation process in the backfill.

The model has been both calibrated and partially verified by laboratory experiments. Models for unsaturated buffer have been evaluated within three international projects (DECOVALEX, CATSIUS CLAY and VALUCLAY). These projects included calculation examples and comparisons with measurement results for both laboratory experiments and field tests. See e.g. /Börgesson and Hernelind, 1995; Alonso and Alcoverro, 1997; Börgesson and Hernelind, 1997; Börgesson et al, 2004/.

In recent years another finite element program has been tested and used for thermal and hydraulic processes, namely Code Bright /Cimne, 2002/. This code is developed by UPC. Most processes are similar to the processes in ABAQUS but there are some important differences. The main difference is that it can handle the gas phase and thus also model processes B1, B4, B5, D2, E2 and F1. It can also handle phase transitions (C1 and C2).

Several large-scale field tests have been installed with e.g. the purpose to study the wetting process of the buffer material in a repository.

The big field test BMT (Buffer Mass Test) was performed in the years 1981 to 1985 in Stripa, Sweden. In this test, highly-compacted bentonite and heaters were installed in six simulated deposition holes on a scale of 1:2. The tunnel above two of these holes was backfilled with sand-bentonite mixtures containing 10 and 20 percent bentonite. The tests were interrupted after a few years, whereupon buffer and backfill were excavated with detailed sampling and water ratio determination. The results showed that the wetting had gone to near water saturation in three of the holes, while the others had not increased their water content more than marginally. The water inflow in the three first-mentioned holes was greater than in the others. On the other hand, the gap towards the rock was left open in these three holes, while the gap in the three "dry" holes was filled with bentonite powder. There was no tendency towards uneven water uptake from the rock. The three wet holes with empty gaps had a high, uniform water ratio with a degree of saturation higher than about 80 percent. The three dry holes with powder-filled gaps were dry nearest the heater and wet nearest the rock, in a way that could be expected to result from the redistribution process that occurs in the presence of a temperature gradient.

In Äspö HRL several deposition hole tests with canister and buffer material are running but no test has so far been interrupted and excavated. These tests are the Canister Retrieval Test (CRT), the Prototype Repository and the Temperature Buffer Test (TBT). All tests simulate heating and wetting of the buffer. CRT and TBT have artificial wetting of the buffer through filters covering all or parts of the rock surface, while the Prototype Repository has natural wetting from the rock. All six deposition holes in the Prototype Repository also have an upper boundary of backfill material that contributes to the wetting.

The wetting of the buffer in these tests have been modelled and, for the Prototype Repository, the influence of the rock structure and properties have been studied in detail /Börgesson and Hernelind, 1999/. This study was originally done for the safety analysis but has also been used for the Prototype Repository. It is applicable for deposition holes in general. In the study the permeability of the rock matrix, the water supply from the backfill, the water pressure in the surrounding rock, the permeability of the disturbed zone around the deposition hole, the water retention properties of the rock, and the transmissivity of two fractures intersecting the deposition hole have been varied.

The results indicate that the wetting takes about 5 years if the water pressure in the rock is high and if the permeability of the rock is so high that the properties of the bentonite determine the wetting rate. However, it may take considerably more than 30 years if the rock is very tight and the water pressure in the rock is low. The calculations also show that the influence of the rock structure is rather large except for the influence of the transmissivity T of the fractures, which turned out to be insignificant for the values used in the calculations.

The results of the calculations show that the influence of most changes in rock properties and boundary conditions are rather large. There is a lack in knowledge of rock properties and interaction buffer/rock and a need for improved material models of the buffer.

Natural analogues/observations in nature

Wetting of bentonite in nature has not been studied and can probably not contribute to the knowledge.

Time perspective

The above analyses show that the time to full water saturation of the buffer can vary a great deal, depending above all on the interaction with surrounding rock. If conditions are optimal for Swedish rock, saturation may be reached within 10 years. If there are no fractures and no supply of water from the backfill, all water has to go via the rock matrix, which may take 100 years in the granitic rock types occurring in Sweden.

The influence of the backfill material on the wetting of the buffer has recently been investigated in a number of calculations that are not yet reported. The main observations from these calculations are the following:

If wetting is achieved from the backfill (30/70) the time to full saturation of the buffer may be reduced with 50% at normal rock conditions

In the extreme (and not applicable) case that no water at all is supplied by the rock (impervious rock) neither to the buffer nor the backfill all water must be taken from the existing water in the backfill. If 30/70 mixture with 12% water ratio is placed as backfill the water ratio will be reduced to about 11% and the degree of saturation in the buffer will be increased to about 98% in 1000–2000 years.

Another extreme case is that no water is supplied by the rock to the deposition hole but that a fractured zone intersecting the deposition tunnel rather soon yields water saturation and full water pressure 5 MPa in the backfill. This scenario yields a time to saturation of 250–500 years.

Very long time until saturation is probably not a problem but the consequences have not been fully investigated. It can be divided into two cases:

1. No water can leave the deposition hole.
2. Water can leave the deposition hole and enter the backfill.

If water is supplied to the backfill or if the backfill has a naturally high RH (e.g. 30/70 with the water ratio 12%, which has an intial RH of 97%) the backfill and the swelling buffer in contact with the backfill will create a natural lid that does not allow any water to escape. On the other hand if the backfill is composed of bentonite blocks with low RH, which are piled in the deposition tunnel, and if the deposition tunnel as well as the deposition hole are very dry, there is a possibilty that the hot environment around the canister in combination with slots between the blocks may dry the buffer and lead the moist air out into the backfill by mainly convection.

In case 1 (no water is allowed to leave the deposition hole) the consequence will only be an internal redistribution of water and attendant salt accumulation in the hot parts. This case is treated in chapter 4.7.

Case 2 combined with a slow water inflow into the lower part of the buffer may lead to a salt accomulation in the buffer around the canister that is far stronger than if only internal redistribution takes place.

Handling in the safety assessment SR-Can

The initial THM evolution covering the saturation phase of the buffer will be modelled for a number of hydraulic boundary conditions in SR-Can since the supply of water at the outer buffer boundary is decisive for the wetting rate. Calculations will be made both with the old THM-model used for SR-Can /Börgesson and Hernelind, 1999/ and updated models, e.g. with Code Bright.

The system evolution model /Hedin, 2004/ will be used to study the swelling of the buffer and the compaction of the backfill at the end of the saturation phase for different assumptions regarding critical input data like friction and groundwater salinity. This additional study is done since one of the critical results of the swelling at the end of the saturation phase is the redistribution of buffer mass leading to a reduction of buffer density at the top of the deposition hole.

Time scales: The THM modelling will encompass the wetting period for the backfill and buffer. The detailed time scale will consequently be determined by the hydraulic conditions in the bedrock, which control the wetting. Typical time scales range from less than ten to more than a hundred years.

Boundary conditions: The THM modelling will be performed for a number of different hydraulic conditions in the bedrock.

Influences and couplings: The process is coupled to a number of other processes in the THM model. The modelling will be done for a realistic thermal gradient over the buffer and include thermal expansion, the swelling of the buffer at the end of the saturation process (considering friction between buffer and the wall of the deposition hole), the associated compaction of the backfill and possible movements of the canister in the deposition hole as a consequence of these phenomena. All variable influences in the table above will be included in the modelling except effects of varying pore water composition where in particular the salinity of the pore water could influence the final swelling of the buffer. The latter is however included in the system evolution model where the salinity effect on swelling is implemented according to section 2.7.8 (osmosis).

The special cases of failed canister and of earthquakes: Canister failures and earthquakes of a magnitude that could affect the wetting process are not expected during the time period in which the buffer saturates. The process is thus not handled for these cases. Should the assumptions regarding significant, early canister failures or earth quakes not be confirmed by the results of the safety assessment, the handling of the special cases will be reconsidered.

Uncertainties

The knowledge of the hydraulic behaviour of unsaturated buffer material is gained from investigations done on MX-80 while almost no tests have been performed on the other buffer candidate (Milos bentonite). At the density considered for the buffer material the behaviour is similar but this needs to be confirmed.

Uncertainties in mechanistic understanding

The understanding of the process is deemed sufficient for the needs of the safety assessment. Some hydraulic parameters and processes in the buffer are uncertain e.g. the suction close to water saturation and the influence of the swelling pressure (currently studied), the influence of entrapped air and the magnitude of vapour flux and the influence of gaps, which imply very inhomogeneous initial conditions. Although detailed knowledge of these processes might not be needed for the safety assessment they will be studied in order to gain further confidence in the wetting modelling.

Model simplification uncertainties for the above handling in SR-Can

The water uptake modelling done so far has assumed that the bentonite blocks and the slots (filled or unfilled) are homogeneous so that the buffer has the same density everywhere from start. This simplification is not correct but the effect of the error is likely to be smaller than that of the uncertainty of the rock properties.

The detailed description of the wetting process is complicated, but it can be modelled with sufficient accuracy for the needs of the safety assessment for which it is not important to know all the details of the process. It is however important to show that full water saturation is eventually reached in order to provide a reliable description of the long-term evolution following saturation and to determine the distribution of buffer and backfill mass at the end of the saturation phase. It is further important to assess the possible chemical alterations of the buffer during the saturation process. Such alterations are however not expected to influence the saturation process and need hence not be considered for this process.

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

The greatest uncertainty/variability concerns the hydraulic conditions in the near-field rock and the hydraulic interaction between the rock and the buffer. This will be addressed by analysing a number of cases with differing assumptions regarding these properties.

2.3.2 Water transport under saturated conditions

Overview/general description

Water transport in saturated buffer is a complex interplay between several sub-processes on a microscopic scale. On a macroscopic level, the result is that the permeability of a saturated buffer is very low, and this is also the essential result for the safety assessment.

Water flow in saturated buffer is a special case of unsaturated flow. The processes involved are the same as for unsaturated conditions, but can vary widely in importance compared with unsaturated conditions.

The most important mechanism under saturated conditions is transport of water in liquid phase, which is driven by a water pressure gradient. The process can be described by Darcy's law. Any deviations from Darcy's law, which can occur at low gradients, are favourable in that they lead to a reduced through flow rate. The hydraulic conductivity K is a function of the composition of the buffer, the void ratio e, the ion concentration in the pore water i_c and the temperature T.

The hydraulic conductivity of the reference buffer (MX-80 and Milos) to low-salinity water after complete saturation is approximately 10^{-13} m/s at the density 2000 kg/m^3 at water saturation.

An approximate condition for transport by flow to be more important than diffusive transport is given by: $D_e/\Delta L < Ki$, where D_e = the effective diffusivity, ΔL = the transport length (one-dimensional), K = the hydraulic conductivity and i = the hydraulic gradient. For the stage after restoration of the hydraulic situation in the repository, i can be set equal to 10^{-2} and, for anions $D_e = 10^{-12}$ m^2/s, which for the buffer thickness $\Delta L = 0.35$ m requires that K exceeds $3 \cdot 10^{-10}$. This is at least two orders of magnitude higher than the real conductivity of the buffer. The margin for cations, which have a lower diffusivity, is considerably greater. The conclusion is thus that the only important transport mechanism for both water and dissolved species through the buffer is diffusion. Diffusive transport is dealt with in section 2.7.3.

The demand for having a hydraulic conductivity that is so low that advection (flow) can be neglected has led to a demand of the hydralic coinductivity of the buffer of

$K<10^{-12}$ m/s,

which is about 2 orders of magnitude lower than theoretically required. Since this demand can only be fulfilled if all slots are healed and there is a perfect contact between the buffer and the rock a demand for the swelling pressure σ_s is also made:

$\sigma_s >1$ MPa

Influencing/influenced variables

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	Yes	
Water content	No	Yes	Influenced via density
Gas content	Yes (of solved gases)	No	
Hydrovariables (pressure and flows)	Yes	Yes	
Buffer geometry	No	Yes	
Pore geometry	No	Yes	
Stress state	No	No	
Montmorillonite composition	No	Yes	
Pore water composition	Yes	Yes	
Bentonite composition	No	Yes	
Structural and stray materials	No	Yes (insignificant)	

Influence of density, temperature and salinity:

The hydraulic conductivity is primarily dependent on the geometry and composition of the buffer, the density, the ion concentration in the pore water and the temperature.

In the buffer, $K = 5 \cdot 10^{-14}$–$5 \cdot 10^{-13}$ m/s at the densities in question both for MX-80 and Milos bentonite. The dependency on density is rather strong. Figures 2-2 and 2-3 show the hydraulic conductivity of MX-80 and Milos bentonites measured at different densities and with different ion concentration in the saturating water.

Transformations to other minerals with reduced montmorillonite content as a result generally leads to a higher hydraulic conductivity. The temperature influences the conductivity by changing the viscosity of the water, whereby a temperature increase from 20°C to 90°C entails an approximate increase in conductivity by a factor of 3. The influence of salinity on the hydraulic conductivity is at buffer densities not very strong. Investigations are ongoing as shown in Figures 2-2 and 2-3.

Boundary conditions

The outer boundary conditions are determined by the hydraulic properties of the rock surrounding the deposition hole, in particular the properties of possible fractures intersecting the deposition hole and of the excavation damaged zones that may exist in the peripheries of both deposition holes and deposition tunnel floor.

Model studies/experimental studies

Flow in water-saturated buffer has only been studied on a laboratory scale in the form of permeability tests and swelling/compression tests in oedometers /Börgesson et al, 1995/.

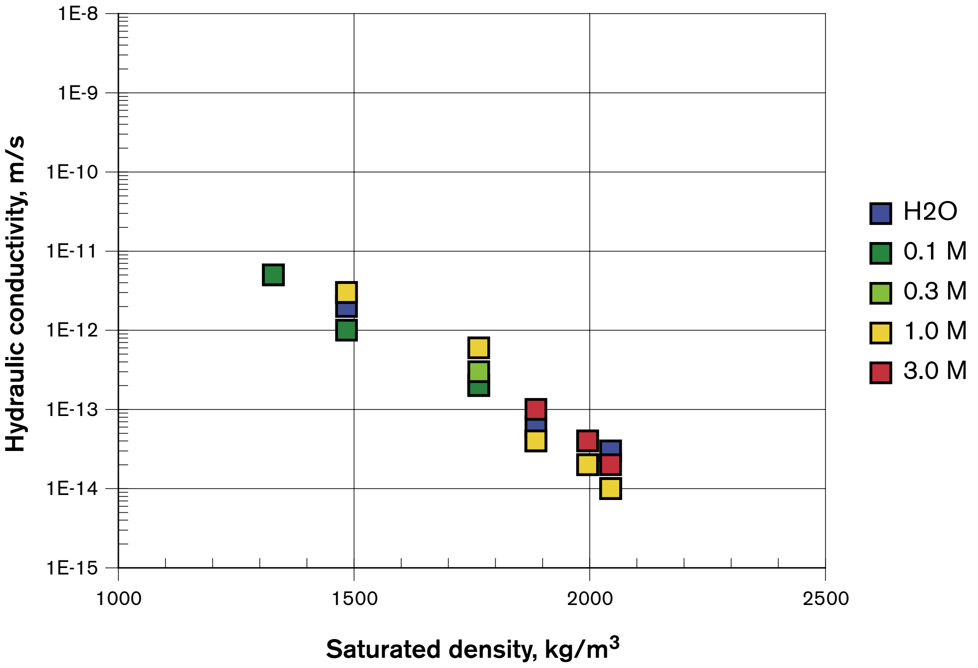


Figure 2-2. Hydraulic conductivity of MX-80 bentonite measured at different densities and molar concentration of NaCl in the saturating solution.

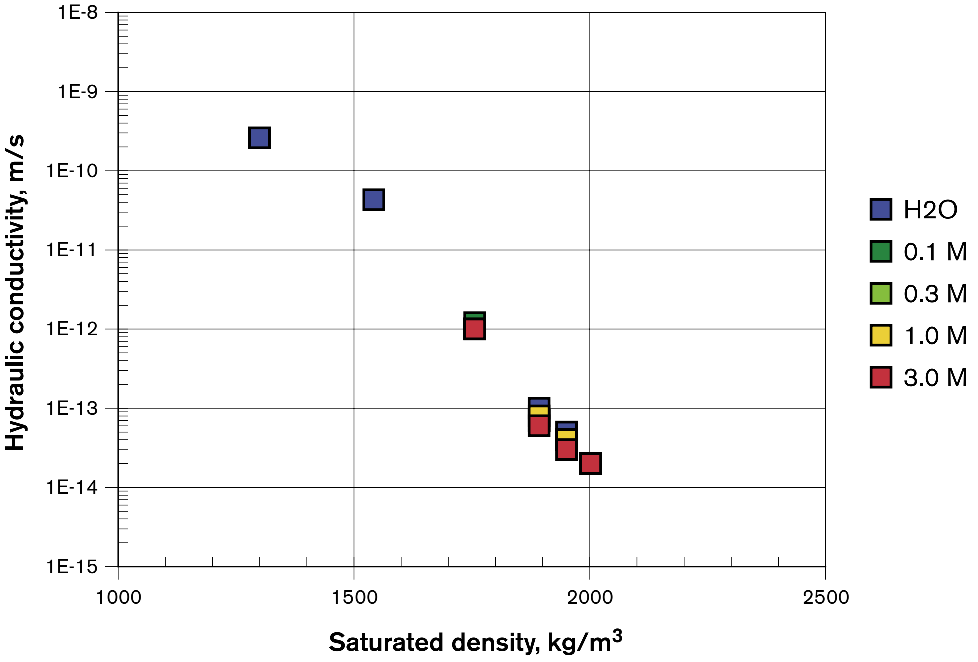


Figure 2-3. Hydraulic conductivity of Milos bentonite measured at different densities and molar concentration of $CaCl_2$ in the saturating solution.

Natural analogues/observations in nature

No studies of water flow in natural buffer like formations have been done.

Time perspective

The hydraulic gradient that exists after water saturation and pore pressure equilibrium have been reached around a repository is expected to be low. A simple estimate can be made of the time required for all the water in the buffer to be exchanged. If the gradient is set to 0.01, the hydraulic conductivity K to 10^{-13} m/s and the cross-sectional area to 10 m^2, according to Darcy's law it takes 15 million years for the approximately 5 m^3 of water present in the buffer to be exchanged.

Handling in the safety assessment SR-Can

The primary handling of this process will be to consider how the hydraulic conductivity could vary for the evolving conditions in the buffer throughout the assessment period. If the hydraulic conductivity is below 10^{-12} m/s, and the swelling pressure above 1 MPa, the process will be neglected.

Time periods: After saturation and throughout the assessment period.

Influences and couplings: The evolution of buffer density, temperature and ionic composition will be studied and the hydraulic conductivity and the swelling pressure will be evaluated for the changing conditions caused by the chemical evolution, section 2.7.5 and alterations of density, section 2.6.1. The conductivity will be evaluated by application of the empirical relationships described above. The swelling pressure will be evaluated according to sections 2.6.1 and 2.7.8.

Boundary conditions: The hydraulic gradient for altered conditions during permafrost and glaciation will be estimated.

The special cases of failed canister and of earthquakes: These are handled via possible mass redistributions, see section 2.6.1. Pressure transients need possibly to be considered for earthquakes.

Uncertainties

So far very few tests have been performed on Milos bentonite but those results indicate that the properties are very similar to the properties of MX-80 at the densities considered for the buffer material (see Figures 2-2 and 2-3).

Uncertainties in mechanistic understanding

An uncertainty exists as to whether Darcy's law applies at the low gradients that are expected to arise, since all laboratory tests are done at very high gradients. All measured deviations show, however, that the flow is lower than is expected by Darcy's law (see e.g. /Dixon et al, 1999/).

Another uncertainty is the effect of transformations, which for some cases are expected to give higher hydraulic conductivity. The effects of very high salinities are also inadequately understood.

Model simplification uncertainties for the above handling in SR-Can

The model used is empirical, connecting hydraulic conductivity to temperature, density and ionic strength of the pore water. The uncertainties in this description are primarily given by the uncertainties of the empirical relationships on which the model is based. The primary aim is to establish that the transport through diffusion is considerably more important than advective transport, meaning that the exact value of the hydraulic conductivity is a secondary concern.

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

Input data for the study of hydraulic conductivity are primarily the empirical relationships between hydraulic conductivity and density, temperature and pore water composition described above. The primary aim is to establish that the transport through diffusion is considerably more important than advective transport, meaning that the exact value of the hydraulic conductivity is a secondary concern.

2.3.3 Gas transport/dissolution

Overview/general description

Transport of gas in the buffer can occur in two phases of the repository's evolution:

- When the repository is sealed, air will be trapped in the buffer. As the buffer becomes saturated with water, the air must escape.
- If a canister should be defective such that water could penetrate through the copper shell, the cast iron insert is expected to corrode under hydrogen gas formation. If more hydrogen is produced than can be dissolved in the water in the canister, a gas phase will form.

Gas which is trapped in or by the buffer can escape by two principal mechanisms:

- If the production rate is low or the gas quantity small, the gas can be dissolved in the pore water and be removed by diffusion.
- If the production rate is high or the gas quantity is large, a gas phase will form, the pressure will rise, and a flow path is expected to be formed through the buffer at a critical pressure.

Water saturation phase: Gas flux in conjunction with wetting of the buffer is described in section 2.5.1. Under these conditions, all gas is expected to be dissolved in the buffer's pore water and transported by diffusion.

Gas transport from failed canister before saturation: Before the buffer is saturated, water is only expected to be able to get into an assumed damaged canister via vapour-phase diffusion. Hydrogen production from corrosion will therefore be limited and the gas that is formed is expected to be able to leave the buffer. The gas transport resistance in an unsaturated buffer is also considerably lower than in a saturated one. (Canister damages at this early stage are very unlikely.)

Gas from defective canister after saturation:

Diffusion:

Hydrogen gas formed by corrosion of the cast iron insert can dissolve in the pore water and migrate from the canister by diffusion. The maximum transport capacity for different conditions has been estimated by /Wikramaratna et al, 1993/.

The results of the calculations show that the diffusive transport capacity is considerably lower than the hydrogen gas production from corrosion, if it is assumed that the entire surface of the cast iron insert is accessible for corrosion and the water supply does not limit corrosion. Under such conditions it is probable that a gas phase will be formed inside the canister and that the gas must escape by gas-phase flow.

Gas flow:

The following picture of the subsequent course of events is an interpretation of the results of theoretical and experimental studies: At a critical pressure (entry pressure), the buffer is expected to allow the gas to pass through. When the pressure reaches this value, a transport pathway is formed through the buffer and gas is released. If gas production is maintained, the pressure will fall to a steady-state pressure. When the production ceases the pressure falls, and if the gas production is low enough, the transport pathway is expected to close. This takes place at a so-called "shut-in pressure", which is dependent on the swelling pressure. At pressures lower than the shut-in pressure, gas migrates solely by diffusion.

Gas entry, breakthrough, peak and steady-state gas pressures in the buffer are dependent on the properties of the buffer. The most important parameters seem to be the swelling pressure, the mechanical properties of the buffer and the stress situation.

Based on the experimental interpretation by /Harrington and Horseman, 2003/, at the peak pressure a fracture will open and the peak pressure could be expressed as:

$$p_g\ (\text{peak}) = T + 2\sigma_{eff} + p_w$$

according to the 'classic theory' of hydrofracture /Haimson and Fairhurst, 1967/, where T is the tensile strength of the clay, σ_{eff} is the isotropic effective stress outside the region of stress concentration and p_w is the porewater pressure in the clay.

In the post-peak region, the bentonite will have lost its tensile strength normal to the plane of the fracture. The gas pressure is now equal to the total stress, σ. On this basis, the gas pressure at any point on the post-peak transient can be written as:

$$p_g\ (\text{post} - \text{peak}) = \sigma = \sigma_{eff} + p_w$$

At the (near) steady-state condition, the gas permeability of the fracture network must remain more or less constant with time, with no tendency for crack dilation or closure. Conceptually, the fracture must be propped open at any point along its length by the local gas pressure.

When gas production ceases, gas pressure drops and then gradually falls away along a very protracted pressure transient. The gas pressure and total stress are approximately equal at all points along the transient.

As gas pressure decreases, individual flow pathways should collapse leading to declining overall gas permeability. The gas pressure cannot fall below the sum of the capillary pressure, p_c, and the externally-applied water pressure (backpressure), p_{we}, or:

$$p_g\ (\text{shut - in}) = p_c + p_{we}$$

where p_c is the capillary pressure, which is equal to the swelling pressure. This provides a lower bound value for the gas pressure at shut-in.

The energy that drives the gas transport comes from compression of the gas inside the canister and is proportional to the available gas volume. If it is assumed that the volume is a cubic metre and the gas pressure drops from 12 to 9 MPa, 30 MJ will be released when the transport pathway is formed. In a system with confined clay (deposition holes), the gas will presumably be released in a controlled fashion, i.e. it will not entrain the buffer material along with it, but experiments under open conditions, i.e. without surrounding counter-pressure, show that gas breakthrough can be a violent process /Donohew et al, 1998/.

Available experimental results show that gas can migrate through a highly-compacted buffer without jeopardizing the continued function of the engineered barriers. However, no experiment has been conducted with a gas volume equivalent to the volume in the canister cavity (approximately 1 m^3), and it is therefore not completely clear what effect the release of large quantities of gas will have on the performance of the repository.

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	Yes	
Water content	Yes	Yes	
Gas content	Yes	Yes	
Hydrovariables (pressure and flows)	Yes	Yes	
Buffer geometry	No	Yes	*It is important to demonstrate that the gas does not influence the Buffer geometry
Pore geometry	No	No	
Stress state	No	Yes	
Bentonite composition	No	No*	Indirect influence through Stress state
Montmorillonite composition	No	No*	Indirect influence through Stress state
Pore water composition	No	No*	Indirect influence through Stress state
Structural and stray materials	No	No	

The gas transport resistance in an unsaturated buffer is considerably lower than in a saturated one. Tests on unsaturated Avonlea bentonite have shown breakthrough pressure to be negligible below a degree of saturation of around 80 to 90% /Hume, 1999/. This range marks the saturation at which there are no continuous gas channels through the clay. In Kunigel VI bentonite, the critical saturation for interconnected gas voids was found to be 72% /Tanai et al, 1997/.

Boundary conditions

The essential boundary conditions for this process are the geometries of the conducting features at the buffer interfaces to canister and rock around the deposition hole and the pressure and volume of gas trapped inside the inner interface. The geometrical aspects of the boundaries include the opening of a defective canister, fractures intersecting the deposition hole and excavation damaged zones around deposition hole and in the floor of the deposition tunnel.

When the gas has reached the excavation-disturbed zone (EDZ) in the near-field rock, the pressure required to get it to migrate further is much lower than in the buffer. It must exceed the sum of the water pressure and the capillary tensions in the fine fractures in the EDZ or in channels in fractures that intersect the deposition holes, which together gives a pressure of 5–10 MPa.

Model studies/experimental studies

Gas dissolution: No specific studies have been conducted for the purpose of studying how gas dissolves in the pore water in the bentonite. However, experience from water saturation tests shows that highly-compacted bentonite is normally completely water-saturated and that no trapped gas remains. The mechanistic interpretation is that the swelling pressure of the bentonite compresses trapped gas, which is dissolved in the pore water and transported out by diffusion.

Gas transport:

A number of gas migration experiments in compacted clays, with different materials, geometries and boundary conditions, have been performed over the last 20 years. Several of these are summarized in /Harrington and Horseman, 2003/.

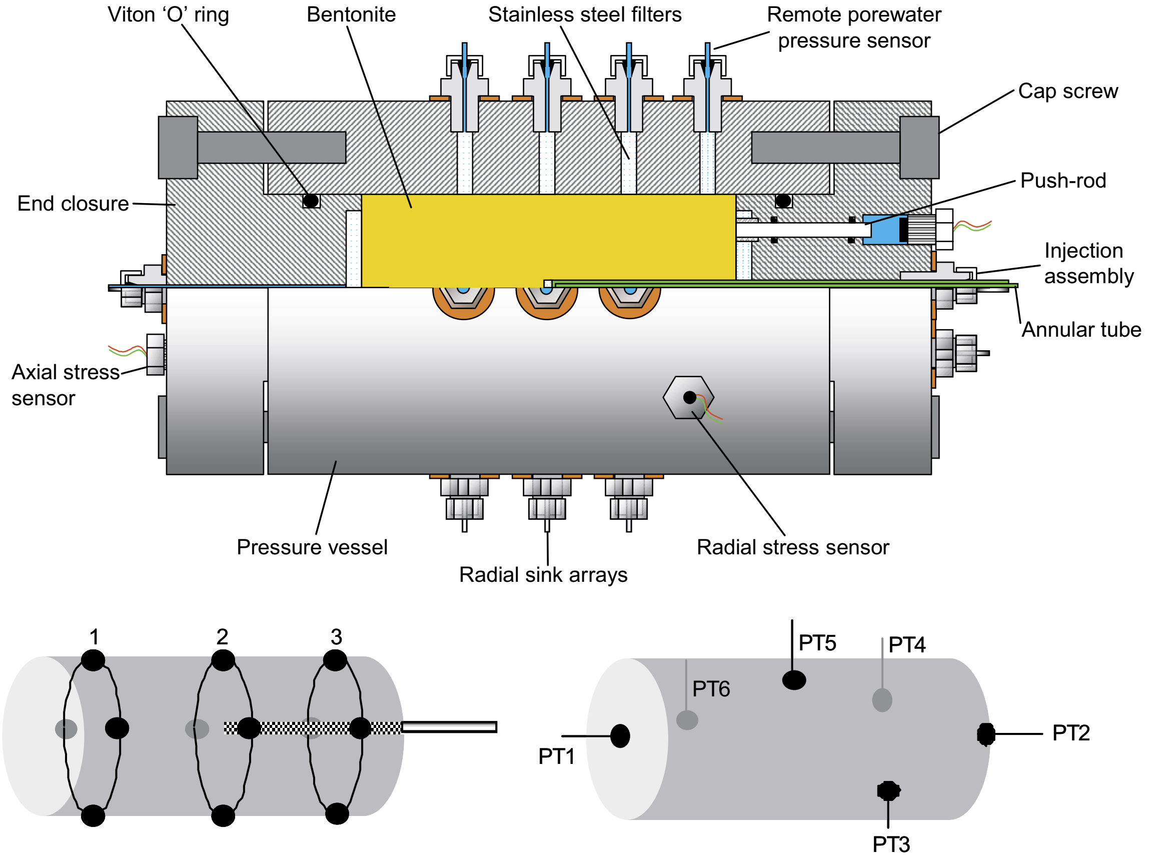
/Harrington and Horseman, 2003/ presents the most recent gas migration experiments within the SKB programme. In the test geometry, the specimen was volumetrically constrained, preventing dilation of the clay in any direction. Figure 2-4 shows the actual test set up.

All bentonite specimens were fully-hydrated before gas testing by backpressuring using de-aired and distilled water. This procedure gave initial degrees of saturation which were demonstrably close to 100%. Figure 2-5 shows the gas injection history from one of the tests.

Gas testing began by pumping gas into the upstream system at a rate of 375 $\mu l \cdot hr^{-1}$. All filters, other than the central source filter and the radial sink filters, were closed off during gas testing. A very small amount of flow was observed at a gas pressure of 13.8 MPa, accompanied by a slight increase in axial and radial total stress. Gas pressure continued to rise until major gas entry occurred at a gas pressure of 18.9 MPa. Injection gas pressure momentarily dropped to 18.7 MPa and there was a sharp rise in axial total stress from 1.6 MPa to 18.3 MPa. Gas pressure then continued to rise to a peak value of 19.4 MPa, at which point large quantities of gas began to flow to sink array [1]. It would appear that the first gas pathway did not intersect a sink and a short period of growth was required in order to obtain gas breakthrough.

It should be noted that the measured peak pressures in these constant volume test are substantially higher than the ones measured in the earlier constant stress experiments.

There is strong evidence that gas flows through a network of pressure-induced pathways. Very little, if any, displacement of water occurs during gas movement. The crack-like pathways are propped open by the elevated gas pressure. When gas pressure falls, individual pathways can snap shut leading to a reduction in overall gas permeability. What is absolutely clear from these gas injection experiments is that it is possible to pass up to 60 litres (STP) of helium through specimens of buffer clay (at gas pressures of over 8.0 MP for times in excess of 5 months) without any measurable desaturation occurring. This strongly reinforces the observations made by others on the lack of desaturation occurring during gas flow through fully-hydrated bentonite.



Sink configuration

Stress sensor locations

Figure 2-4. *Cut-away diagram of the constant volume and radial flow (CVRF) gas migration apparatus. Sensors are as follows: [PT1] – axial total stress on the backpressure end-closure, [PT2] – axial total stress on the injection end-closure, [PT3] – radial total stress close to the injection end-closure, [PT4] – porewater pressure close to the injection end-closure, [PT5] – radial total stress at the mid-plane, and [PT6] – radial total stress close to the backpressure end-closure.*

There is no evidence from these tests that the development of pressure-induced gas pathways in any way compromises the sealing capacity of the bentonite barrier. Gas pathways are ephemeral features of the buffer which tend to close up when gas pressure falls. Breaks in slope of monitored pressure transients can be interpreted as discrete pathway 'sealing' events which coincide with decreases in the rate of discharge of gas to the sinks. More importantly, a history of gas movement through the buffer has been shown to have no detectable effect on permeability to water.

A consortium of radioactive waste disposal agencies, the GAMBIT Club, has been supporting efforts to develop an appropriate model of gas migration through bentonite, with the objectives that the model (a) should adequately represent the principal features observed in experiments of gas migration through compacted bentonite, (b) can be used to analyse and interpret experimental results, and (c) will provide the basis of a model that can be used to assess pressure build up and escape of hydrogen gas through bentonite around waste canisters in a repository /Rodwell et al, 2003 (MRS)/. From that study a number of conclusions can be drawn: A model in which gas migration in highly compacted bentonite occurs by fissure propagation is able to account qualitatively for the behaviour seen, although heuristic arguments are required to explain the dependence of the gas permeability on gas pressure after breakthrough, and the model is limited in the range of phenomena it portrays.

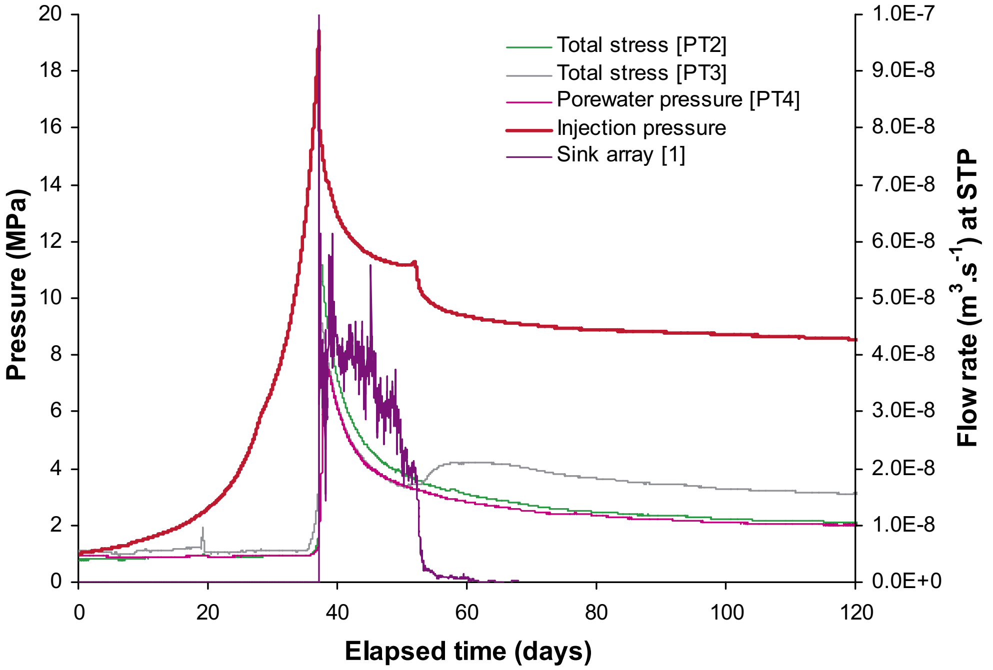


Figure 2-5. *Gas injection pressure versus time for the first breakthrough and shut-in stages of test Mx80-8, showing axial and radial total stress, porewater pressure and flow rate from sink array [1]. Flows to sink arrays [2] and [3] were too small to be distinguished when plotted on this scale. Mass balance considerations show that the flow from sink array [1] is predominantly gas.*

a) To explain the observed experimental results on the assumption that gas displaces water from capillary-like pathways, with the threshold for gas entry controlled by the capillary entry pressure, requires that the capillary radii (or pore throats) be very small to give rise to the large threshold entry pressures reported for highly compacted bentonite, but that a large number of pathways must then become gas filled to give rise to the observed gas permeability. While such circumstances may be possible, they would be unexpected.

b) The continuum model of gas pathway propagation with deformation of the clay does describe a range of observed phenomena, including pathway resealing and hysteresis through changes in the water content of the clay, although the agreement obtained with experimental data is only of comparable quality to that obtained with the other models.

c) A model that extends a formulation analogous to that referred to in the previous paragraph to include a proper treatment of the interaction of stresses and strains in the clay with gas migration gives comparable agreement in simulations of the same experiment. Work to simulate gas propagation in samples of different geometrical configurations is on-going.

Experimental data on the mechanism of gas migration in bentonite to allow confirmation of the appropriate choice of modelling approach are currently lacking.

Natural analogues/observations in nature

/Tissot and Pelet, 1971/ discuss oil and gas movements in clay shale: “The extraction of oil or gas from a finely structured clay matrix runs contrary to the capillary laws and is in principle impossible. However, the barrier can be broken in one way. The pressure in the fluids that is formed in the pores in the clay increases when kerogen is formed. When this pressure exceeds the mechanical strength of the clay, microchannels will form which are orders of magnitude larger than the natural pores and will thereby allow an oil or gas phase to pass, until the pressure falls below a threshold value and the channels fill again and a new cycle begins." These observations recall the conclusions from the gas transport experiments in bentonite.

Time perspective

Gas transport can occur when water is in contact with the cast iron insert in a damaged canister.

Handling in the safety assessment SR-Can

Unsaturated conditions: The gas quantity in a deposition hole is approximately six percent of the whole volume at atmospheric pressure. When the hydrostatic pressure has been built up, the gas volume will have declined by a factor of 50. This small quantity is expected to rapidly dissolve in the pore water and can be neglected in the water saturation calculations.

Saturated conditions: The bentonite buffer is an important barrier in the KBS-3 system. The key purpose of the buffer is to serve as a diffusional barrier between the canister and the groundwater in the rock. An important performance requirement on the buffer material is to not cause any harm to the other barriers. Gas build-up from corrosion of the iron insert could potentially affect the buffer performance in three ways:

1. Permanent pathways in the buffer could form at gas break-through. This could potentially lead to a loss of the diffusional barrier.
2. If the buffer does not let the gas through, the pressure could lead to mechanical damage of the other barriers.
3. The gas could de-hydrate the buffer.
4. A gas phase could push water with radionuclides through the buffer.

The process is neglected except for deposition holes containing a defective canister. For that case, results of model and experimental studies of gas build-up and transport must be considered in an integrated assessment of the internal evolution of a damaged canister, including corrosion of the cast iron insert and water transport in the canister.

The global rate of corrosion of the insert determines if the hydrogen gas produced can escape by diffusion or if a separate gas phase will be formed. The corrosion rate also determines the rate of pressure increase. This pressure will be transferred to the near-field rock and the backfill. At a certain gas pressure, the buffer will open and let the gas through. This maxium pressure is (probably) dependent on the swelling pressure of the buffer. It is also influenced by the hydrostatic pressure and therefore the effect of a glacial overburden, leading to increased pressures, needs special consideration. The gas pathway in the buffer will stay open as long as the gas production rate is sufficiently high.

The integrated assessment of the internal evolution of a damaged canister will be carried out in the SR-Can Main Rerport.

After repository closure, the only direct flammability hazard to man that could arise is from the release of hydrogen at the surface. This has been assessed and found to be extremely unlikely to present any hazard (the calculation was for release into a building /Baker et al, 1997/). The flammaibilty hazard will not be analyzed in SR-Can.

Uncertainties

Uncertainties in mechanistic understanding

Gas transport: A remaining uncertainty in the understanding of gas transport in the buffer material concerns the number, size and spatial arrangement of the gas-bearing fractures and the volume behaviour of the clay during gas injection. As mentioned in the Experiments/Models section above, the observed gas transport through bentonite can be interpreted in different ways.

One critical uncertainty is the break-through pressure, i.e. the pressure when the buffer opens and lets the gas through. This determines the maximum pressure that can be created within the near-field of the repository. Another uncertainty is the closure pressure, the pressure where the pathways in the bentonite do close.

These uncertainties will be considered in the integrated assessment mentioned above.

Model simplification uncertainties for the above handling in SR-Can

The evolution of the gas pressure and the gas transport in bentonite in SR-Can is still entirely based on experimental results. The uncertainties regarding the interpretation of the experiments make predictive modelling difficult at this stage (see above).

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

The maximum gas pressure in the near-field is determined by the break-through pressure in the bentonite. In some experiments, this pressure has been found to be above 20 MPa for a bentonite with a swelling pressure of ~6 MPa. The high pressure may be an effect of the small experimental specimen, but since no data from large scale experiments are available, high breakthrough pressures cannot be ruled out.

Also the gas production (corrosion) rate, which determines the time scale of the gas evolution history, is uncertain.

These data uncertainties will be considered in the integrated assessment mentioned above.

2.3.4 Piping/erosion

Overview/general description

Water inflow into the deposition hole will take place mainly through fractures and will contribute to the wetting of the buffer. However, if the inflow is localized to fractures that carry more water than the swelling bentonite can adsorb, there will be a water pressure in the fracture acting on the buffer. Since the swelling bentonite is initially a gel, which increases it density with time as the water goes deeper into the bentonite, the gel may be too soft to stop the water inflow. The results may be piping in the bentonite, formation of a channel and a continuing water flow and erosion of soft bentonite gel. There will be a competition between the swelling rate of the bentonite and the flow and erosion rate of the buffer.

Piping will take place if the following two conditions are fulfilled:

1. The water pressure p_{wf} in the fracture, when the water flow is prevented, must be higher than the sum of the counteracting total pressure from the clay and the shear resistance of the clay.

2. The hydraulic conductivity of the clay must be so low that water flow into the clay is sufficiently stopped to keep the water pressure at p_{wf}

Erosion will take place if the drag force on the clay particle from the water movement is higher than the sum of the friction and attraction forces between the particle and the clay structure.

Piping probably only occurs before complete water saturation and homogenisation since the swelling pressure of the buffer material is very high. Erosion can occur both as a consequence of channels caused by piping and after long time in the interface between the clay and the fractures in the rock. Since the water flow rate in the latter case is very low erosion will only be important for colloids leaving the clay gel that has penetrated into the fractures, see further section 2.7.10. Piping and erosion are thus two completely different processes, but in order for erosion to take place there must be an open channel caused by piping (except for the colloid case).

If piping has occurred it is essential for the function of the buffer that the buffer swells and closes the channel. The ability of the buffer to seal the channel requires the following

1. The mass transport rate of the swelling of the bentonite into the channel must be larger than the mass transport rate of the erosion
2. When the channel has been sealed piping must not occur again (see above).

Influencing/influenced variables

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	No	
Water content	No	Yes	1)
Gas content	No	No	
Hydrovariables (pressure and flows)	Yes	Yes	
Buffer geometry	Yes	Yes	2)
Pore geometry	Yes	Yes	
Stress state	Yes	Yes	
Montmorillonite composition	No	Yes	4)
Pore water composition	No	Yes	3)
Bentonite composition	No	Yes	4)
Structural and stray materials	No	No	

1) A change in water content changes the swelling pressure, which influences the piping risk. It also influences the hydraulic conductivity, which influences the swelling rate.

2) The distance between the bentonite blocks and the rock surface strongly influences the disposition to piping, since it affects the time to reach a high swelling pressure

3) The salinity of the water affects many variables that are important for disposion to piping and erosion, i.e. the swelling pressure, the swelling rate (through hydraulic conductivity) and the erodability

4) The bentonite and montmorillonite composition also affects variables that are important (see 3)

Boundary conditions

The piping and erosion that are treated here only takes place at the interface between the buffer material and the rock.

Model studies/experimental studies

The processes are studied in conjunction with studies for the KBS-3H concept[1], which is particularly vulnerable to erosion of buffer material since a channel can pass a large number of canisters. Tests have been done in three different scales. The results show that the bentonite is very sensitive to piping and erosion and that it may take a considerable time until it heals if the inflow in one spot from a fracture is strong and the built-up of water pressure in the fracture is fast. These results have not been reported yet.

One of the deposition holes in the Prototype Repository has a rather large water inflow and may be used as a check of one scenario for piping erosions effects. However, this section of the test is not planned to be excavated until after at least another 10 years.

Natural analogues/observations in nature

Not applicable for this process.

Time perspective

The piping and erosion (excluding colloid erosion) only takes place before completed saturation and homogenisation. The high swelling pressure of the buffer will ultimately always seal the piping channels, since the sum of the swelling pressure and the pore water pressure in the buffer will always be higher than the water pressure. The water inflow must though not be so strong that the buffer has lost too much bentonite through erosion.

Handling in the safety assessment SR-Can

The maximum consequences of this process will be estimated by model calculations. This will be based on the experiences from the KBS-3H study.

Time periods: The process is only relevant during saturation and neglected for all other time periods.

Boundary conditions: The hydraulic boundary conditions at the buffer/deposition hole interface are decisive for this process.

Influences and couplings: The consequence of an extensive piping together with erosion would be a loss of buffer density.

The special cases of failed canister and of earthquakes: The process only occurs at very early stages when these special cases are not relevant.

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[1] In the KBS-3H concept, canisters are deposited horizontally in long tunnels, rather than vertically in individual deposition holes.

Uncertainties

Piping, erosion and subsequent sealing is a complicated process with many components, much depending on the hydraulic behaviour of the rock. The uncertainties are considerable regarding both the demands on the rock and the ability of the buffer. For the KBS-3H concept, a preliminary demand is that one canister position must be able to handle the inflow 0.1 l/min and a pressure increase of the water pressure in a fracture intersecting the tunnel of 100 kPa increase per hour. This could thus be transferred to demands of the deposition hole, but it is not obvious that the demands should be equal. Furthermore, the time for the buffer to seal such an inflow from a fracture is not known.

2.4 Mechanical processes

2.4.1 Swelling/mass redistribution

The swelling process has been merged with other processes that cause mass redistribution within the buffer, namely thermal expansion, creep and a number of interactions with the canister, near field rock and the backfill.

Overview/general description

Water is absorbed by both unsaturated and saturated bentonite and causes swelling. If the bentonite is unable to expand freely, a swelling pressure develops, which locally reaches its peak at full water saturation.

In a deposition hole the buffer is inhomogeneous from start due the slots at the horizontal buffer boundaries. When water from the rock fills the outer, pellets filled, slot and enters the bentonite blocks there will be swelling of the blocks and compression of the pellets filled outer slot. At first the swelling will be strong since the density of the pellets filled slot is low and the resistance against compression thus small. This means that the outer part of the blocks will swell to a lower density than the average density expected after complete homogenisation before the water has been sucked so deeply into the blocks that the swelling pressure compresses both the pellets part and the swelled outer part of the blocks. With time after completed saturation the compression of the outer part and the expansion of the inner parts will come to some kind of equilibrium, which (due to inner friction in the bentonite and hysteresis effects) will not be complete homogenisation. Instead a remaining small density gradient with higher density close to the canister and lower close to the rock will endure.

The swelling also leads to compression of the backfill above the deposition holes, a mechanical pressure on the canister and rock surface of the deposition hole, and clay intrusion in fractures around the deposition hole. Depending on the conditions in the near-field rock, the clay in the fractures could be carried away by the groundwater, allowing new clay to penetrate, which could lead to gradual erosion of the buffer (see section 2.7.10). Aside from these mechanical effects, the buffer's hydraulic conductivity and diffusion properties are altered by the swelling.

Other phenomena that could lead to mass redistribution, expansion or contraction of the buffer include creep, shear movements and convergence of the deposition hole, canister movements, pressure exerted by canister corrosion products and thermal expansion of the buffer pore water.

The following description illustrates the principles for modelling of swelling from a geotechnical perspective and does not concern the underlying mechanisms.

The swelling can be conceived of as being caused by a force of repulsion between the montmorillonite layers. If there is a limited supply of water in a free specimen, the swelling is counteracted by a negative pressure in the pore water. If a specimen is water-saturated, i.e. all pores are filled with water, the swelling is counteracted by the formation of a negative pressure in the pore water in the water menisci on the surface of the specimen. The negative pore pressure is equal to the internal swelling pressure if no external pressure is applied. If the specimen is unsaturated, the water menisci develop inside the specimen as well. The negative pressure in the pore water is chiefly a function of the water ratio in the specimen, i.e. the quantity of water per unit weight of dry material. This negative pressure is called suction potential (see also section 2.5.1). When water is added to a free specimen, the water ratio increases, and the repulsion forces and the suction potential decrease, which causes the specimen to swell until a new equilibrium is established with a lower internal swelling pressure. If the volume is kept constant, a portion of the internal swelling pressure is instead transferred to an external swelling pressure, which can be measured. When a specimen with constant volume is completely water-saturated and the pore water pressure is kept positive, the entire swelling pressure becomes an external pressure. At water saturation, the swelling pressure and the pore water pressure are independent quantities and give a total pressure that is the sum of the pressures.

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	Yes (insignificant)	Yes	Through thermal expansion
Water content	Yes	Yes	
Gas content	Yes	Yes	Degree of saturation
Hydrovariables (pressure and flows)	Yes	Yes	
Buffer geometry	Yes	Yes	1)
Pore geometry	Yes	Yes	
Stress state	Yes	Yes	
Montmorillonite composition	No	Yes	Adsorbed cation species
Pore water composition	No	Yes	
Bentonite composition	No	Yes	
Structural and stray materials	No	Yes	

1) Any phenomenon leading to alterations of the buffer bulk geometry influences swelling; see further “Boundary conditions” below.

The data and discussion on influences in the following text are in principle valid for MX-80 but the mechanical properties of Milos bentonite is similar to the properties of MX-80, which means that they can in large also be applied to Milos bentonite.

Measured swelling pressure at constant volume after completed water saturation is shown in Figures 2-6 and 2-7 for both MX-80 and Milos bentonites. The influence of density and ion concentration in the saturating water is measured and included in the figures.

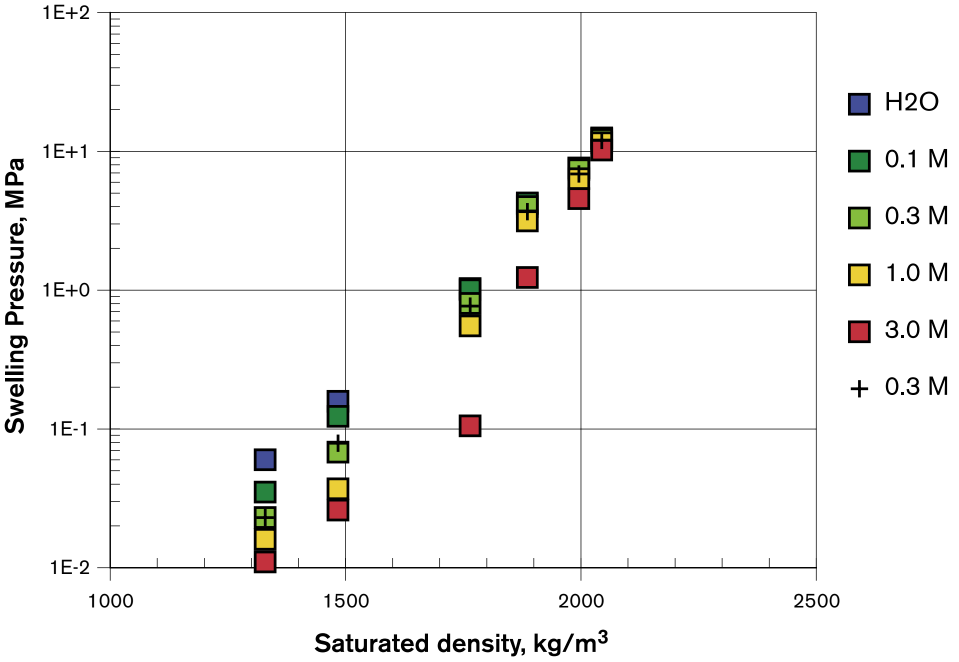

Figure 2-6. *Swelling pressure of MX-80 bentonite measured at different densities and molar concentration of NaCl in the saturating solution*

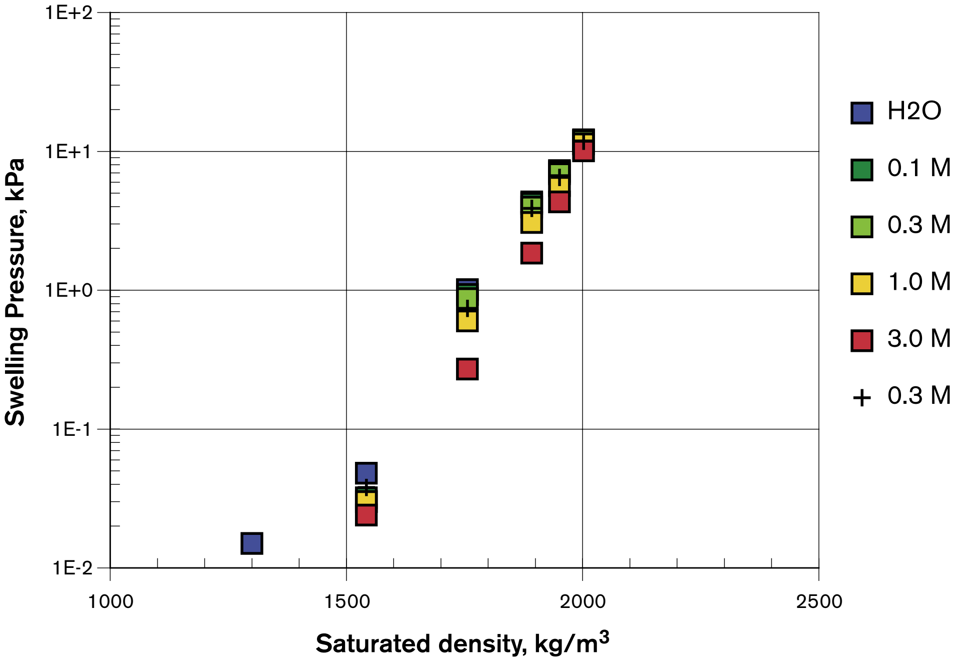

Figure 2-7. *Swelling pressure of Milos bentonite measured at different densities and molar concentration of $CaCl_2$ in the saturating solution*

Influence of density: The density at water saturation is decisive for the water ratio, which determines the swelling pressure. Figures 2-6 and 2-7 illustrate the difference in swelling pressure at different densities.

Influence of montmorillonite content and species of adsorbed cation: According to Figures 2-6 and 2-7, the swelling pressure is relatively independent of the adsorbed ion is sodium or calcium if the density exceeds approximately 1,800 kg/m^3 and the montmorillonite content are the same. At lower densities the swelling pressure is lower if the adsorbed ion species is Ca. Reduced montmorillonite content can be conceived of as being equivalent to the addition of an inactive aggregate.

Influence of chemical composition of pore water: Increased ion concentration generally leads to a reduced potential swelling, and thereby to effects on the pore geometry that are similar to those of an ion exchange to divalent ions. Increased ion concentration further leads to a reduction of the swelling pressure due to osmotic effects, and to changes in the pore geometry that affect the hydraulic conductivity of the material at low buffer densities (see section 2.7.8).

Influence of degree of saturation: In an unsaturated state, the bentonite/water system shrinks on drying and expands on wetting. The volume change on water uptake in e.g. a block is less in the unsaturated state, since the swelling takes place partially in previously unfilled pores. Shrinking due to drying is expected to take place near the canister, which can lead to cracking of the buffer. After full saturation the cracks will be healed and the buffer homogenised in the same way as all initial slots and joints between blocks in a deposition hole.

If volume is restrained during water uptake, which is the case for the buffer in a deposition hole, the expansion will be replaced by a swelling pressure that gradually increases as the degree of saturation increases.

Influence of temperature: A change in the temperature of the buffer will cause water, particles and pore gas in the buffer to expand and may influence the mechanical properties of the buffer.

The influence of temperature on the mechanical properties of the buffer in the water-saturated state is small /Börgesson et al, 1995/. In the unsaturated state, this influence is presumably small /Villar, 2002/ but is not sufficiently well-known for MX-80.

The expansion of the particles and the pore gas is relatively small. The expansion of the water, which is about 100 times greater than the expansion of the particles, completely dominates this process.

The influence of the water's expansion is completely dependent on whether the buffer is water-saturated or not. Before water saturation the volume of the water can expand with virtually no other resistance than that offered by the compression properties of the pore gas. At constant volume this leads (to simplify slightly) to an increase in the degree of saturation when the temperature increases. The consequences of this are that the negative pressure in the pore water (suction potential) decreases and that the total pressure and the pore gas pressure increase. This causes the swelling pressure against an external restraint to increase slightly. At constant pressure, a slight increase of the volume is obtained in the same way. However, these changes are small and apparently without practical significance. The increase in degree of saturation is about 2 percent at a temperature increase of 50 degrees.

The consequences can be more serious in water-saturated buffer, since there are no gas-filled pores to swallow the volume increase. At constant volume the pore pressure increase is great, since the water must be compressed. Theoretically, the resultant pressure increase against the surroundings can amount to several tens of MPa at the temperature changes in question. However, the pressure increase is counteracted by drainage of the water through the rock and backfill. This causes a decrease in pore pressure, which occurs in parallell with the temperature increase. This process is relatively fast and can greatly reduce the pore pressure increase if the temperature increase is slow.

Boundary conditions

The boundary conditions of interest mainly concern alterations of the geometry of the buffer's interfaces to the near field rock, to the canister and to the deposition tunnel backfill.

Interaction buffer/backfill

In the interface between the buffer and the backfill, the buffer exerts a swelling pressure against the backfill and vice versa. Since the difference in swelling pressure may be great, a net pressure arises against the backfill whereby the buffer swells and the backfill is compressed. In this process, the swelling pressure from the buffer decreases as the density decreases. At the same time, the counter-pressure from the backfill increases as it is compressed and its density increases. The swelling of the buffer and compression of the backfill are counteracted to some extent by friction against the rock. When the force of the swelling pressure in the buffer is equal to the sum of the force of the counter-pressure in the backfill and the friction against the rock, the process ceases since equilibrium has been established.

The size of the swelling depends on the original densities of the buffer and the backfill and associated expansion and compression properties.

Due to the friction against the rock and friction in the buffer, the homogenization of the buffer is not complete. The density reduction due to the swelling therefore decreases with increasing distance from the interface. For typical densities in the backfill (30/70), the density reduction of the buffer probably does not reach down to the canister, which lies 1.5 m below the interface.

Figure 2-8 illustrates the displacement of the interface between the buffer and backfill and the displacement of the canister as a result of a finite element calculation of the wetting of the buffer for a rock with one fracture intersecting the centre of the deposition hole. /Börgesson and Hernelind, 1999/.

Interaction buffer/near field rock

The following mechanical interactions between buffer and near-field rock have been identified:

1. **Swelling pressure from the buffer** is transferred to the rock but is not expected to lead to significant rock movements. The elastic deformation of the rock at the expected swelling pressures is small due to the rock's high modulus of elasticity. Plastic deformations caused by movements along a fracture plane can occur along rock wedges that go up to the tunnel floor.
2. **Pressure from the canister** can be propagated via the buffer. Such pressures can arise from canister corrosion products and gas pressure build-up.
3. **Forces of friction** arise against the rock walls in the deposition hole due to swelling of the buffer against the backfill.

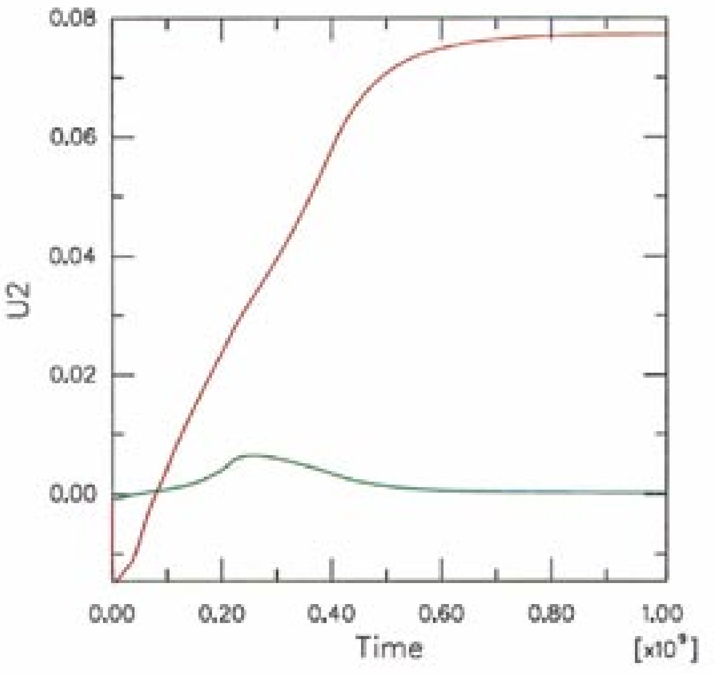

Figure 2-8. *Displacement (m) of the buffer/backfill interface (red) and the canister (green) as a function of time (s) elapsed from deposition.*

4. **Thermal expansion of the buffer** can be serious for the rock if the buffer is water-saturated when the temperature increase occurs. This process is described above. The slow temperature increase in combination with the drainage through the rock implies that this is not a problem but it should be checked by some calculations.

5. **Convergence of the deposition hole** occurs when the rock creeps due to the high rock stresses after rock excavation. Assuming an anisotropic primary state of stress with reasonable components, calculations have shown that the diameter can decrease by approximately a centimetre after 10,000 years /Pusch, 1995b/. The resulting density increase in the buffer leads to an increase in the swelling pressure by roughly a few MPa. Recent conservative estimates of creep-induced deformations of deposition tunnels have given convergence values of about 1% /Glamheden et al, 2004/. For a deposition hole this would give between 1 or 2 cm of diameter reduction. The recent estimates are based upon the notion that creep will relax fracture shear stresses over time until a low threshold stress has been reached. The effect can be even greater at higher rock stresses, caused by e.g. glaciation.

6. **Shear movements due to sliding** along a fracture in the rock is a process that can be serious for the canister if the displacement is rapid and great. The process is partially dealt with below (Interaction buffer/canister). Rock movements are dealt with in the forthcoming process descriptions for the geosphere, see the Process report for SR 97 /SKB, 1999/ for an earlier description.

7. **Intrusion of bentonite into fractures in the rock** is very limited due to the small aperture of the fractures and the shear resistance caused by the friction between the bentonite and the fracture surface. If there is no loss in bentonite due to erosion the effect is positive since it implies a sealing of the fracture. Problems may occur if the gel that forms at the outer part of the penetrated bentonit is eroded and carried away by the ground water.

The depth of the intrusion is a function of the fracture width, the friction between the bentonite and the rock surface and the swelling pressure in the deposition hole. The penetration depth z can be calculated according to the following equation (derived from simple integration of force equilibrium):

$$z = \frac{\delta}{2\tan\phi}\left(\ln\sigma_0 - \ln\sigma_z\right)$$

where

δ = fracture width

φ = friction angle (10–30 degrees)

σ_0 = swelling pressure on the rock (7–13 MPa)

σ_z = swelling pressure in the fracture at the distance z from the rock surface

The penetration depth will thus go towards infinity when the swelling pressure at the gel limit approaches 0. If the swelling pressure 1 kPa is set as a limit the penetration from a buffer with the swelling pressure 10 MPa into a fracture with the width 0.1 mm will be 1.3 mm if the friction angle 20 degrees is assumed (an average). If the limit pressure 1 Pa is set the penetration will be 2.2 mm. Extrapolating the swelling pressure relations (not verified) one finds that 1 kPa corresponds to the void ratio e=70 (w=2500%) for fresh water and e=3.5 (w=125%) for sea water.

At the salt content 0.6%, which is relevant for Swedish repository conditions, the maximum swelling capacity of MX-80 is swelling to a water ratio of 215% /Wold, 2003/. This corresponds to a density at saturation of 1 250 kg/m^3 At a horizontal fracture of 0.1 mm width the effective stress on the "bottom" of the fracture from the own weight of the gel will thus be 0.25 Pa, which yields an average effective stress of 0.125 Pa that the gel never can pass. If this limit is applied the penetration will be limited to 2.5 mm.

Since the penetration depth is proportional to the fracture width a rule of thumb may be that the penetration is limited to 25 times the fracture width.

This process is also dealt with in section 2.7.10.

General comments

The influence of the buffer on the rock is difficult to investigate experimentally in any other way than by measuring the swelling pressure on the rock. This has been done in BMT in Stripa /Pusch et al, 1985a/ and in several of the full-scale tests in Äspö HRL /Goudarzi et al, 2003/. Calculations of the effect on the rock of corrosion of the cast iron insert show that the influence is little /Bond et al, 1997/. The effect on the rock of the buffer's upward swelling in the deposition hole has been modelled /Pusch and Börgesson, 1992/. These studies show that a rock wedge can, if the backfill is poorly compacted, move upward 0.2–0.3 m and reduce the density of the buffer in the upper part of the deposition hole. This scenario presumes that a triangular wedge reaches from the floor in the tunnel more than a meter down to the buffer in the deposition hole. It also presumes that the wedge is attached to the rock during installation of the buffer and backfill but detached either during compaction of the backfill or due to thermal stresses (or similar) after deposition. The swelling pressure and the axial upwards swelling of the buffer can than move the wedge upwards and create an increased volume of the deposition hole.

The influence of the rock on the buffer in connection with shear movements along fracture planes has been studied in both experimental and model studies. This process is not harmful for the buffer (if its rheological properties have not been changed) but only for the canister (see below).

Processes 1, 3, 4 and 7 occur during the first 100 years after closure of the repository. The other processes occur or can occur throughout the lifetime of the repository.

The uncertainties concerning processes 1–4 and 6 stem mainly from uncertainties in other processes and are dealt with elsewhere (canister corrosion, gas formation, canister/buffer interaction, etc).

Convergence of the deposition holes is normally negligible.

The penetration of buffer into fractures is of no importance for the buffer unless followed by erosion. This process is treated in section 2.7.10.

Interaction buffer/canister

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

The mechanical interaction between buffer and canister is of the following nature:

1. Swelling pressure and water pressure exerted on the canister.

The magnitude of the swelling pressure is described above. During the saturation process the swelling pressure may be uneven mainly due to uneven wetting, but after full saturation the swelling pressure is rather homogeneous. The water pressure is added to the swelling pressure and is also homogeneous with the expected magnitude given by the depth of the repository. During glaciation the water pressure may be substantially higher.

2. Thermally induced pore water pressure exerted on the canister.

This process is described above. The process can be dangerous for the canister if the following two criteria are fulfilled:

1. The buffer is water saturated during increasing temperature
2. The water volume decrease caused by the pressure reduction during drainage of the water through the buffer out to the rock and backfill is slower than the water volume increase caused by the thermal expansion of the water during the increasing temperature.

Preliminary calculations have shown that the latter criterion is not probable /Pusch and Börgesson, 1992/. The former criteria may be fulfilled but is not enough on its own for causing high pore pressure.

3. Trapped gas in the buffer exerts pressure on the canister.

Gas generation is treated in section 2.5.3. High gas pressures are only expected as a possible consequence of corrosion of the insert of a failed canister. In that case, the gas pressure will act on both the inside and the outside of the canister and the net mechanical effect on the canister should thus be negligible.

4. Canister corrosion products exert pressure on the canister.

External corrosion of the copper canister and corrosion of the steel insert in case of a defective copper canister, will increase the volume of the canister since the corrosion product has a lower density than the original material. This material expansion will compress and consolidate the buffer and thus increase the pressure from the buffer on the canister. This process will be further treated in the forthcoming reporting of canister processes.

5. Vertical movement of canister in buffer.

The average density of the canister is much higher than that of the buffer, which means that there is a net vertical stress on the bottom part of the buffer in the deposition hole. This stress difference results in a density that is slightly higher below the canister than in the other parts of the buffer. This means that there is a net vertical sinking of the canister in relation to the original location. On the other hand, there is an upward swelling of the buffer above the canister (due to the compression of the backfill; see above), resulting in a decreased density and decreased swelling pressure above the canister. This may lead to a swelling pressure below the canister that is higher than the sum of the weight of the canister divided by the cross sectional area and the swelling pressure on the top of the canister. Since the upward- directed force on the canister thus will be higher than the downward-directed force, the canister will heave instead of sink.

There is thus a complicated combination of pressure (and forces) on the canister. The result can be calculated and leads to equilibrium after complete water saturation and pore water pressure stabilisation, which includes stress differences and shear stresses in the buffer. This equilibrium is typically established within 100 years after closure of the repository whereupon creep phenomena will dominate the displacements in the buffer.

There are three types of time dependant displacements of the buffer resulting in movements of the canister. One is related to what in soil mechanics is named “primary consolidation”, the second one to so called “secondary consolidation” (volumetric creep) and the third to time dependant shear deformations (deviatoric creep).

During primary consolidation the pore water pressure is the regulating variable and the time perspective is determined by the hydraulic conductivity and the compression properties. Before equilibrium in effective stresses in the clay skeleton is established, the pore water carries the total pressure difference. With time, as the pore water pressure difference is reduced due to outflow of water, the pressure is transferred from the pore water to the clay skeleton and the clay is thus deformed. This process dominates as long as there are pore water pressure differences in the buffer.

After pore pressure equilibrium the creep processes dominate. The volumetric creep is caused by internal microstructural reorganization of the clay particles and is related to the change in swelling pressure with time. This creep is not particularly strong and not problematic as far as vertical movements of the canister are concerned, since it will not remove any bentonite but (in the case of constant stress) only compress it or (in the case of constant volume) relax the stresses.

The deviatoric creep is potentially problematic since the existence of deviatoric stresses (or shear stresses) is dependant on internal friction or cohesion in the clay. These parameters are slightly reduced with time, which causes a deviatoric creep and thus a movement of the canister. If the friction and cohesion are reduced to zero the clay would behave as a viscous liquid and there would be no resistance against the canister sinking to the floor of the deposition hole.

The deviatoric creep has been investigated with a number of creep tests at different stresses and a creep model has been derived /Börgesson et al, 1995/. The model has been used in preliminary finite element calculations. The calculations were actually made on the concept with two canisters in a deposition hole but the results are also applicable for the reference concept with one canister. They show that the creep settlement after water saturation is a few mm in 300 years and less than a cm if extrapolated to 100,000 years /Börgesson, 1993b/.

Further studies of deviatoric creep will aim at establishing a critical buffer density, such that if the density exceeds the critical value, canister sinking can be ruled out.

6. Transfer of shear stresses from rock via buffer to canister, especially during an earthquake

An earthquake may induce a shear displacement along a fracture that intersects a deposition hole. The rock displacement will pressurise the bentonite in the deposition hole on the active side and cause high shear stresses that lead to plastication in the area between the fracture and the canister. The pressure on the active side will be transferred to the canister and make the canister partly tilt and partly be bent.

The effect on the canister is depending on the stiffness or shear strength of the bentonite in relation to the stiffness of the canister. The shear strength of bentonite is dependant on the density and the rate of deformation. This scenario has been thoroughly investigated recently and is described under "Model studies" below.

7. Intrusion of buffer in a damaged canister

Bentonite may penetrate into a damaged canister. The influence on the buffer is though small due to the small void available inside the gap between the copper and the insert.

Model studies/experimental studies

Saturated bentonite

Bentonite's swelling capacity and swelling pressure under water-saturated conditions has been studied in detail by means of laboratory experiments /Börgesson et al, 1995/. The results have been used to devise a material model that can be used for FEM calculations /Börgesson et al, 1995/. It is based on the effective stress theory and coincides largely with models for other soils, although the magnitude of the swelling and pressure are considerably greater for bentonite.

The material models have been used to calculate different evolutions that are associated with volume changes in a repository. Examples of modellings are the following:

- Swelling of buffer against the backfill in deposition holes /Pusch and Börgesson, 1992/. A calculation of an extreme case was performed for PASS /Pusch and Börgesson, 1992/ where the buffer was assumed to have a density of 2.1 t/m^3 after water saturation and the density of the backfill was assumed to be 1.9 t/m^3 after water saturation, which corresponds to higher density in the buffer and lower in the backfill than is stipulated in the reference case /Bäckblom, 1996/. This gave an upward displacement of about 30 cm.

- The thermo-hydro-mechanical calculations of the water saturation phase of a deposition hole with different hydraulic rock properties, done for SR 97, included the displacement of the buffer/backfill interface and the movements of the canister /Börgesson and Hernelind, 1999/. The average density of the buffer after saturation of 2000 kg/m^3 and an E-modulus of the backfill of 30 MPa resulted in a heave of the backfill interface of 7–8 cm and a temporary heave of the canister of about 1mm.
- Settlement experiment in Stripa /Börgesson, 1993a/. Comparisons of calculated and measured movements and densities when canisters are loaded in deposition holes.
- Modelling of canister movements for design with two canisters in a deposition hole /Börgesson, 1993b/. In conjunction with studies of the function of deposition holes with two canisters, a calculation was made of the movements of the canisters which included upswelling towards the backfill /Börgesson, 1993b/. In this calculation the buffer was defined to have a high density (2.1 t/m^3 at saturation) and the backfill was assumed to be rather stiff, with a density after water saturation of 2.2 t/m^3. The calculated upswelling of the buffer was about 7 cm and the total heave of the canister (including both cosolidation and creep) after 32000 years was 3 mm..
- For a rock shear the influence of the density and the rate of rock displacement has been investigated for very fast shearing and reported recently /Börgesson et al, 2004/. The influence is illustrated with the following examples: At the density 2000 kg/m^3 at water saturation the shear strength is 1 MPa at the shear rate $5 \cdot 10^{-8}$ m/s (4 mm/day) while the shear strength is 3 times higher (3 MPa) at the shear rate 1 m/s. At the density 2100 kg/m^3 at water saturation the shear strength is 2.6 MPa at the shear rate $5 \cdot 10^{-8}$ m/s (4 mm/day) while the shear strength is also at this density about 3 times higher (7.6 MPa) at the shear rate 1 m/s.
- A material model has been established and the effect of rock shear on the canister and buffer calculated. The influence of density, shear rate, magnitude of shear displacement and location of the shear plane has been investigated with finite element modelling /Börgesson et al, 2004/. The results show that there may be strong plastication both of the copper canister and the steel insert but that the plastic strain is limited to a few percent at the reference case (density 2000 kg/m^3, shear rate 0.1 m/s, shear displacement 10 cm).

The maximum plastic strain in the steel insert, after a shear displacement of 10 cm at the density 2100 kg/m^3, is about 9%, which does not yield rupture of the steel but should anyhow be considered an upper limit. The buffer material should thus not have a density higher than 2100 kg/m^3 in order to avoid damages of the canister in case of an earth quake.

Experience from measurements is available from field tests in Stripa. In BMT (Buffer Mass Test), six deposition holes were simulated on half scale. The movement of the interface was measured with a coin that was placed in the interface and levelled before and after the tests. The measured upwards swelling varied between 4 and 7 cm, depending on test time and degree of saturation in the buffer.

Experience is at present also gained from several of the full-scale tests in Äspö HRL. The Canister Retrieval Test is expected to be fully water saturated in the major parts of the buffer at excavation, which is planned to take place 2007.

Several scale tests in the laboratory (mostly in the scale 1:10) have been run to complete water saturation. Two examples:

- Rock shear was simulated in the scale 1:10 in three laboratory tests performed 1986 /Börgesson, 1986/. Before the shear was simulated the bentonite was brought to complete water saturation through filters in the periphery of the buffer and the swelling pressure measured. The three tests were identical with exception of the shear rate that was varied between 0.03 mm/min. to 160 mm/min. The bentonite was carefully sampled and the water ratio and density of the samples were determined after finished test. The tests gave valuable information about both the effect of shear at different rates and the homogenisation of the bentonite in a deposition hole.
- The KBS-3H concept has been simulated in the laboratory also in the scale 1:10. Two canisters with buffer material, perforated steel containers and distance plugs were installed in a tube and mainly the swelling pressure and relative humidity RH were measured. By measuring RH the total suction of the clay and the resulting degree of saturation can be evaluated.. The bentonite was brought to complete water saturation through filters with an increased water pressure of 1 MPa. The test arrangement was left for one year and the axial hydraulic function of the buffer was tested by applying hydraulic gradients in the filters. Finally the test was terminated and water ratio and density measured after comprehensive sampling.

No heating was applied to the two latter test types.

Unsaturated bentonite: A few experiments have been conducted to study the volume change as a function of degree of saturation and void ratio. These experiments have been used to calibrate the material model for unsaturated clay.

The interaction between external stress, pore water pressure (suction) and degree of saturation in a unsaturated bentonite is investigated in a PhD work that has been running for some years and will be finished 2004. This work will shed further light on the development of swelling pressure in an unsaturated buffer with limited volume.

Additional experiments need to be done to improve the understanding and the model.

Thermal expansion has been simulated under water-saturated conditions in laboratory tests, which show that expected stress increases occur, followed by falling stresses due to drainage of water. A numerical modelling of the process /Börgesson, 1992/ shows that the maximum pore pressure increase amounts to 8 MPa due to the fact that the rock drains the buffer. However, this calculation assumes that the rock acts as a filter for the pore water meaning that the pore water is allowed to enter the rock matrix freely.

Time perspective

The water saturation phase is expected to take between 10 and 100 years, depending on the properties of the rock. During this time span, the portion of the expansion that is limited by the consolidation rate (which is controlled by the hydraulic conductivity) probably also takes place. The subsequent volume creep can proceed for thousands of years, but is not judged to be of such a magnitude that it appreciably influences buffer performance. The deviatoric creep will also continue during the lifetime of the repository and mainly affect the canister sinking (see above) but also the homogenisation of the buffer. As mentioned earlier the friction in the bentonite is counteracting the homogenisation and is responsible for e.g. the following effects:

1. The decrease in buffer density at the buffer/backfill interface is not spread to the entire buffer

2. The penetration of bentonite into the fractures in the rock is limited to about 25 times the fracture aperture
3. There will be a remaining radial difference in density originating from the swelling of the blocks and the compression of the pellets

The deviatoric creep can be regarded as a decrease in friction with time. This process, which has been investigated in relation to the canister sinking, is very slow and not expected to have a dramatic influence on the buffer in the lifetime of a repository.

Thermal expansion: Temperature calculations /Thunvik and Braester, 1991/ show that the maximum temperature in the bentonite at the canister wall is reached after about 10 years, at the rock wall after about 30 years and furthest away from the canister after another ten years or so. In the most favourable case, water saturation can be reached between the canister and the rock in a few years and in the whole buffer after 10–15 years. In other words, there is a risk that a pressure increase will arise in the interval between 10 and 50 years after deposition. However, the temperature increase is very moderate and slow during this time interval. Furthermore, a temperature decrease takes place simultaneously nearest the canister after about 10 years, which counteracts the effect. As stated above the judgement is that this process is not a problem but it should be verified with some calculations.

Natural analogues/observations in nature

No natural analogues concerning mechanical behaviour have so far been studied. The existence is probably very limited and difficult to evaluate.

Handling in the safety assessment SR-Can

The process is relevant in all time scales, is influenced by a range of boundary conditions/effects and coupled to a number of other processes.

Early effects

The initial swelling is modelled as part of the integrated initial THM evolution of the buffer, see further section 2.5.1. Also thermal expansion and interaction buffer/backfill is included in this modelling.

The system evolution model /Hedin, 2004/ will be used to study the swelling of the buffer and the compaction of the backfill at the end of the saturation phase for different assumptions regarding critical input data like friction and groundwater salinity, see further section 2.5.1.

Long-term effects

Long-term effects of ion exchange and saline groundwater will be evaluated using empirical relationships described above. Effects of salinity will also be modelled by the Donnan model referred to in section 2.7.8. This is implemented in the system evolution model.

Effects of canister corrosion products: Estimates of volumes and thus of the increase in swelling pressure etc, will be carried out as part of an assessment of integrated, long-term mechanical effects. This will include a quantitative treatment of copper corrosion.

Canister sinking: The process is provisionally neglected, based on the above modelling results. A more developed basis for this assumption, based on a buffer density criterion for the neglect of the phenomenon needs to be established. The consequences of canister sinking will be assessed in a residual scenario.

Effects of convergence of deposition hole: These effects will be estimated based on results from ongoing studies as part of discussion of integrated, long-term mechanical effects.

Effects of intrusion into fractures and subsequent colloid formation and dispersion: Neglected as long as the concentration of divalent cations in the groundwater exceeds 1 mM, otherwise the process has to be quantified. See further section 2.7.10.

Failed canister: Effects of gas generation and of penetration of clay into the canister must be considered in the integrated treatment of a failed canister in relevant scenarios.

Earthquakes: The mechanical effects of earthquakes on the buffer will be handled in separate, integrated assessments in the SR-Can main report, encompassing also earthquakes that do not cause canister failure. Generally, it is noted that a shear movement does not alter the volume of a deposition hole significantly.

Uncertainties

Uncertainties in mechanistic understanding

After complete water-saturation, the swelling process is deemed to be sufficiently well-known and the final density distribution in the buffer can be calculated with sufficient accuracy.

In the long term, swelling properties can change due to transformations and cementation, see section 2.7.9 "Montmorillonite transformation". The mechanical consequences of transformations can be assumed to lead a higher hydraulic conductivity and a decrease in swelling pressure.

Cementation effects may cause significant changes in the physical properties of the buffer, since they can produce a material that is brittle and without swelling capacity and thus can crack and open channels and gaps in the buffer. Since this is a chemical process it is treated elsewhere, see sections 2.7.9 and 2.7.6.

The swelling pressure process and the swelling/compression properties at water saturation are rather well known and can be modelled with sufficient accuracy /Börgesson et al, 1995/.

The mechanical function of water-unsaturated bentonite is complicated to model, and the models that exist today are incomplete, especially for modelling of volume change and swelling pressure. Work is ongoing. The mechanical processes before water saturation is not considered important for safety, but anyhow investigated for improving the general understanding of the saturation phase.

The mechanisms of the different interactions (mechanical interaction buffer/backfill, buffer/rock and buffer/canister) are rather well known regarding the final state after saturation and stress equilibrium.

Thermal expansion: The process is well understood for water-saturated bentonite. For water-unsaturated bentonite the thermo-mechanical theory formation is not complete, but the consequences of this process are deemed in this case to be unimportant for safety.

The largest uncertainty beside the behaviour at unsaturated state concerns the creep. Although a creep model exists and has been verified (for deviatoric stress) the validity for long-term effects is difficult to prove.

Model simplification uncertainties for the above handling in SR-Can

See the above heading.

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

Water saturated state: Some uncertainty arises in calculating the size of the swelling pressure after swelling, where the possible deviation is judged to be ± 30 percent, depending on the scatter in swelling pressure measurements and the uncertainty in certain material parameters. Some validation has been done by means of laboratory /Pusch and Adey, 1986/ and field tests /Börgesson, 1993a/, and continued validation is taking place via the field tests in the Äspö HRL and laboratory tests performed and reported recently /Börgesson et al, 1995; Karnland et al, 2002/ and ongoing tests. Other parameters required are rather well known, e.g. the friction angle (which is a function of density) and the compressibility, which have an uncertainty of 10–20%.

Unsaturated state: Since the mechanical processes during the unsaturated state are rather poorly known, the uncertainties are large. These processes are deemed to mainly influence the path the system will take to reach the saturated state and only to an insignificant degree influence the final, saturated state.

2.4.2 Liquefaction

Overview/general description

Liquefaction is a process implying that a stiff material (soil) turns into liquid due to an affect with short duration, see e.g. /Lambe and Whitman, 1969/. It may take place in a loose sand when the pore water pressure is increased either due to a vibration that makes the sand particles float in the pore water (since they tend to go into a higher degree of compaction, but the water temporarily prevents it) or due to a strong upward water flow that releases the effective stresses between the particles (quick sand). It may also take place in clay that has been settled in salty water (forming an open structure with a high water ratio). If the salt is partly washed out by fresh water, the clay structure cannot hold the high amount of water, meaning that the structure collapses when exposed to vibrations.

These two types of liquefaction cannot take place in a bentonite with high density, since the effective stress that holds the clay together is high due to the swelling pressure. However, a similar phenomenon has been observed during compaction of bentonite blocks at very high water ratios. If the bentonite is compacted at a very high stress to a state where the bentonite is completely water saturated, all further increase in stress will be taken by the water and the bentonite will behave like a liquid. This phenomenon has been observed during uniaxial compaction when liquid bentonite has squirted from the mould.

The process requires a very strong impact of pressure and is rather unlikely to occur in a deposition hole. It can probably only occur at an earthquake and requires a reduced volume of the deposition hole due to an increase in rock stress. A combination of factors may lead to an increase in rock stress of about 15 MPa /Bäckblom et al, 2004/. Estimation according to Kirsch /Brady and Brown, 1994/ of the convergence of a deposition hole at such an increase in rock stress may yield a convergence of the hole of about 1 mm. It can not be eliminated that such a convergence may cause liquefaction.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	No	
Water content	No	Yes	
Gas content	No	Yes	
Hydrovariables (pressure and flows)	Yes	Yes	
Buffer geometry	Yes	Yes	
Pore geometry	Yes	Yes	
Stress state	Yes	Yes	
Montmorillonite composition	No	No	
Pore water composition	No	No	
Bentonite composition	No	No	
Structural and stray materials	No	No	

Boundary conditions

The process requires that the rock boundary of the deposition hole is exposed to a sudden pressure increase with the tendency to reduce the volume of the deposition hole.

Model studies/experimental studies

Block compaction tests have shown that the process may occur. Some laboratory tests are running in order to shed further light on the process. The process should, in similarity to the rock shear case, be subjected to an investigation of the worst possible rock response that can result from an earth quake and then followed by investigations of the influence of such a scenario on the buffer. The example above indicates the magnitude but the matter needs to be more investigated.

Natural analogues/observations in nature

Not applicable.

Time perspective

The process may occur anytime during the lifetime of the repository after completed water saturation. The process requires an earthquake.

Handling in the safety assessment SR-Can

The process will be integrated with handling of earthquakes and resulting fracture movements around deposition holes. It may be necessary to undertake some small-scale laboratory experiments to verify the boundaries for this process.

Uncertainties

The process is unlikely to appear and cannot be modelled today. It is very uncertain if the rock may be subjected to such a stress state, but the estimate above indicates that it cannot be completely ruled out. The uncertainties so far mainly originate from the uncertainty of the behaviour of the rock but to some extent also the buffer.

2.5 Chemical processes

2.5.1 Introduction

Bentonite minerals

High quality commercial bentonites normally contain over 80% of montmorillonite, which gives different bentonite products similar properties in many respects. The distribution of accessory minerals may vary within, and especially between different quarries. Typical accessory minerals are feldspars, quartz, cristobalite, gypsum, calcite and pyrite.

The montmorillonite mineral belongs to the smectite group, in which all the minerals have an articulated layer structure. The thickness of an individual mineral layer is around 1 nm (Figure 2-9) and the extension in the two other directions are often several hundred nanometers. Each layer is composed of a central sheet of octahedrally coordinated cations, which on both sides is linked through shared oxygens to sheets of tetrahedrally coordinated cations. Clay minerals of this type are often referred to as a 2:1 layer structures.

By definition of the montmorillonite mineral, the following applies. The octahedral sheet has aluminum as central ion, which is partly substituted principally by Mg. The tetrahedral sheet has silicon as central ion, which partly may be substituted principally by aluminum. The substitutions results in a net negative charge of the montmorillonite layer in the range of 0.4 to 1.2 unit charges per $O_{20}(OH)_4$ -unit, and the octahedral charge is larger than the tetrahedral. The induced negative layer charge is balanced by cations (c) located between the individual layers (interlayer space). A variable number (n) of water molecules may be intercalated between the individual mineral layers.

The montmorillonite ideal formula may consequently be written:

$Si_{8-x}Al_x$	$Al_{4-y}Mg_y(Fe)$	$O_{20}(OH)_4$	$c_{x+y}n(H_2O)$	$(x<y)$ and
tetrahedral	octahedral		interlayer	$0.4<x+y>1.2$

and the structure may schematically be illustrated as in Figure 2-9.

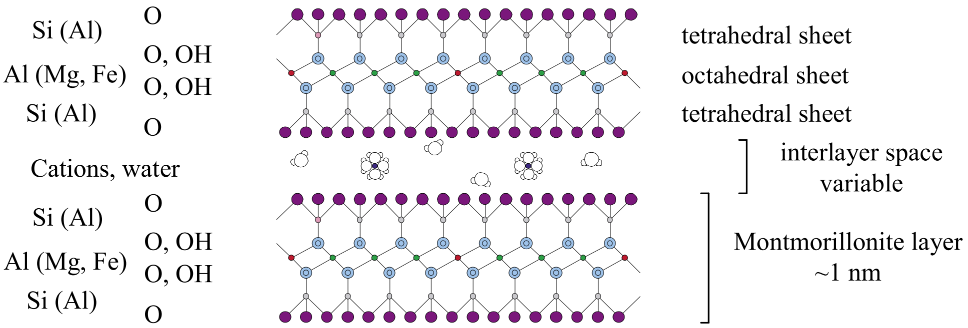

Figure 2-9. *Visualization of two montmorillonite layers showed perpendicular to the mineralogical c-axis.*

Montmorillonite pore-water

Ions in a water solution reduce the water chemical potential, and a concentration difference between solutions normally leads to water transport from the high potential (low concentration) to the low potential volume. In parallel, ions will diffuse from the high concentration to low concentration volume, and the equilibrium condition, i.e. equal chemical potentials, leads to leveled ion concentration.

The total suction of a soil, as described in section 2.6.1, includes osmotic suction and matric suction. By definition, the osmotic suction is due to lowering of the chemical potential in pore-water by ions, and the matric suction refers to the lowering of the chemical potential of the pore-water by mineral surface interaction.

In bentonite, the conditions are extraordinary due to the negatively charged montmorillonite layers and the charge compensating cations in the interlayer space. Water molecules can be intercalated between the individual montmorillonite layers to create an interlayer ionic solution. The cations cannot freely diffuse away from the mineral surface because of the demand for electrical neutrality. Water will consequently be transported into the inter-layer space, if water with a higher chemical potential is available, and the interlayer distance will increase, which is synonymous with bentonite swelling. This water uptake will continue until the chemical potentials equal, which in the case of a pure water source theoretically leads to infinite swelling. The extent of water uptake in bentonite may thereby be orders of magnitude larger and have a different character than in other soil materials.

In a fixed total volume, the water uptake into the interlayer space will reduce the volume of initially larger pores. The uptake is forced to stop when the total available pore volume is completely filled with introduced water. At this full water saturation, water will continue to move in order to level the interlayer ion concentration and thereby the chemical potential in the system. The interlayer distances will thereby increase in interlayer space with high concentration on the expense of interlayer space with low concentration, and the final distances will be a function of the local montmorillonite layer charge. The remaining difference in ion concentration, between the high concentration interlayer solution and the water supplying solution, leads to an osmotic pressure build-up in the clay (swelling pressure), which equals the chemical potential in the system. The homogenization of the pore size, due to the leveling of the chemical potential, may be partly counteracted by mechanically stable structures, e.g. in volumes with complicated stacking of the montmorillonite layers, or in clusters of accessory minerals. In extreme cases the pore-water in such clusters may eventually be in equilibrium with the water supplying solution, without influence from the surrounding montmorillonite charge compensating cations.

The degree of final pore size homogeneity has been analyzed and vividly discussed in the literature, and quite different conceptual and quantitative models have been proposed (ref ??). The conditions are of major importance since the pore-size distribution governs the possible transport of colloids (2.7.4).

The concentration of the charge compensating cations in the interlayer space is highest close to the negatively charged montmorillonite surfaces and will decrease drastically with distance from the surfaces. The distribution, which may ideally be calculated by use of the Poisson-Boltzmann equation, is of major importance for the ion equilibrium with other ions in the system, which may origin from an external solution (2.7.8) or from accessory minerals in the buffer (2.7.6).

The volume and the bentonite mass are fixed in a deposition hole, and the inter-lamellar cation concentration can consequently be calculated. The total pore volume in the buffer in one deposition hole is around 5250 liters, and the bentonite mass is around 19250 kg.

The montmorillonite layer charge may be experimentally determined and are usually given as cation exchange capacity (CEC). The reference bentonites have a CEC of around 0.75 equivalents per kilogram clay material, which give a total cation content of around 14430 equivalents. The mean cation concentration in this system is consequently almost 3 M for monovalent ions if it is in equilibrium with pure water. Figure 2-10 shows the cation distribution in the interlayer for three different interlayer distances at equilibrium with pure water, as calculated by Poisson-Boltzmann equation.

Cementation

The term "cementation" has often been used in a broad sense to describe effects of chemical and mineralogical processes which can lead to various changes in the rheological properties of the buffer material, for example increased mechanical strength, brittleness or reduced swelling capacity. A number of completely different processes could conceivably cause similar effects, and the underlying reactions can occur directly in the montmorillonite mineral, in the inter-lamellar space, or in accessory minerals. The resulting cementation does not necessarily have to appear in the same area as the underlying process, since released species can be transported in the buffer by various prevailing gradients (thermal, concentration, etc) and be precipitated in various locations and in various forms. The chemical processes discussed for the buffer/backfill include those reactions that have been offered as possible causes of cementation of the buffer. It is however not correct to discuss cementation as a process in its own right in this context.

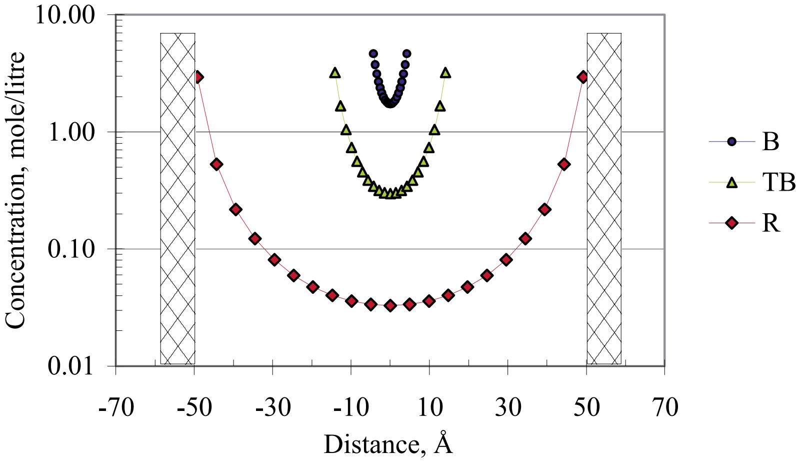

Figure 2-10. *Concentration distribution of charge compensating cations for three different interlayer spacings. B represents mean buffer spacing of around 1 nm, TB represents the mean interlayer spacing in the tunnel backfill material of around 3 nm, and R represents a randomly chosen 10 nm spacing. Two scaled montmorillonite mineral layers are shown to illustrate the geometry for the R-case.*

2.5.2 Advection

Overview/general description

Solutes (dissolved substances) and colloids can be transported with pore water by pressure-induced flow: advection. Hereby, solutes move from areas of higher water pressure to areas of lower pressure. The process leads to redistribution of solutes in the pore water and thus affects the pore water composition. There are several possible causes of pressure gradients in the buffer material, e.g. external water pressure, temperature induced volume change of the water, ion concentration gradients in water, and affinity for water in the bentonite. The advective transport of materia is a direct function of water flow in the buffer, which is comprehensively described in section 2.5.

Advection is of importance in the buffer during the unsaturated period when a net flow of water takes place in the buffer. The principal flow during this phase takes place in the direction towards the canister, provided that the rock supplies the buffer with groundwater. After placement of a canister, a minor flow may initially also take place in the direction from the canister due to the transient temperature gradient.

Under saturated conditions, the transport of solutes in the pore water is expected to be dominated by diffusion, see section 2.5.2. Exceptions may be sudden events such as gas pulses or earthquakes, which can cause local pressure changes in the buffer. The improbable condition with a failure at the bottom of a canister may lead to a partly water-filled canister and hydrogen gas pressure acting on the water. The gas pressure may build up to reach at least the bentonite swelling pressure, which would lead to a significant advective water flow from the canister.

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

Variable/condition	Influencing? a	Influenced? b	Notes
Radiation intensity	No	No	
Temperature	(Yes)	Yes	a) minor b) through water viscosity
Water content	(Yes)	(Yes)	secondary from porosity
Gas content	(Yes)	Yes	a) transport of dissolved gas b) reduced flow in unsaturated buffer
Hydrovariables (pressure and flows)	by definition	by definition	
Buffer geometry	No	1	
Pore geometry	(Yes)	Yes	a) primary in unsaturated clay b) primary by tortuosity
Stress state	Yes	Yes	a) secondary in unsaturated clays b) secondary through pore volume
Bentonite composition	No	No	
Montmorillonite composition	No	No	
Pore water composition	Yes	(Yes)	a) primarily b) especially under unsaturated conditions
Structural and stray materials	No	(Yes)	alternative passage

Boundary conditions

The external boundary condition for this process is the hydrostatic gradient over the buffer.

Model studies/experimental studies

Several studies by SKB and others have studied water flow in bentonite material, both during water uptake and after complete water saturation, and a wealth of quantitative data, which describes water transport is available on different physico-chemical conditions, e.g. densities, temperatures, salt solutions.

The Lot experiments show an increase in total ion concentration in the pore-water compared to what is originally present in the bentonite as a consequence of the salinity of the saturating ground-water. Precipitation of calcite and gypsum close to the central heater indicate ion enrichment (section 2.7.6).

Natural analogues/observations in nature

Comment: Smectite sediments in the oceans keep large pore pressure differences over geological time scales due to the low hydraulic conductivity.

Time perspective

Flow transport in conjunction with water saturation takes place on a timescale of up to hundreds of years. Flow transport after water saturation is expected to be negligible in relation to diffusive transport.

Handling in the safety assessment SR-Can

Before saturation: No detailed modelling of advection. It should though be noted that the composition of the pore water will be influenced by the composition of the intruding water during saturation. If relevant, this should be considered when defining initial conditions for modelling of the chemical evolution after saturation, see further section 2.7.6.

After saturation: Based on the scenario specific evaluation of water transport under saturated conditions, process 2.5.2, advection will normally be neglected compared to diffusion. If the expected low hydraulic conductivity is for some reason not maintained, advection could be included in the modelling of the buffer chemical evolution and of radionuclide transport.

Failed canister: Advective flow from the canister due to hydrogen gas pressure can be calculated as a special case.

Earthquakes: Pressure transients may arise, but will not be prevailing over the buffer over time scales that could lead to significant water flow.

Uncertainties

Uncertainties in mechanistic understanding

See section 2.5.1 for uncertainties concerning water advective transport.

Model simplification uncertainties for the above handling in SR-Can

Unsaturated conditions: see section 2.5.1.

Saturated conditions: The process can be modelled with a standard mass-transfer approach. The main uncertainty lies in the coupling between the fractured rock and the porous buffer.

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

Unsaturated conditions: See section 2.5.1.

2.5.3 Diffusion

Overview/general description

Solutes can be transported in stagnant pore water by diffusion, following concentration gradients. Solutes thereby move from areas of higher concentration to areas of lower concentration. The process leads to a redistribution of dissolved pore water constituents as well as radionuclides in the pore water. Therefore, diffusion is a critical process for radionuclide migration in the buffer, but also affects the actual pore water composition.

The diffusion process is strongly coupled to nearly all chemical processes in the buffer, since it accounts for transport of reactants to and reaction products from the processes. Diffusion of Na^+ and Ca^{2+} ions is, for example, of crucial importance to ion exchange processes, diffusion of K^+ is an important limiting factor for the illitization process, etc. Diffusion is thereby a central process for the entire chemical evolution in the buffer.

Diffusion in bentonite has been thoroughly studied in conjunction with radionuclide transport. Diffusion equations for radionuclides (as well as some experimental measurement techniques relevant for determining the respective parameters) are described in detail in /Yu and Neretnieks, 1997/.

Diffusion in a porous medium takes place according to Fick's first law:

$J = -D_p \varepsilon \, dC_p/dx$

where J is the diffusive flux, D_p the diffusion coefficient in the pore solution, ε is the porosity and C_p the concentration of the diffusing species.

The pore diffusion coefficient in a solution-filled pore is lower than the diffusion coefficient in an unconfined volume of the same solution, D_w. This is mainly due to the tortuosity of the pores, which increases the length of the diffusion path.The relationship between D_p and D_w is:

$D_p = D_w \, \delta/\tau^2$

where δ is the physical constrictivity and τ the tortuosity of the pores. The constrictivity takes into account any narrowing or widening of pores along the flow path. Since bentonite is viewed as a homogeneous porous medium on a macroscopic level, δ is ususally set equal to unity.

Fick's second law, taking into account conservation of mass, describes changes in concentration of a diffusing species in time and space. For diffusion of a sorbing species in a porous material, it can be formulated as follows:

$$\varepsilon\, \partial C_p/\partial t + \partial q/\partial t = \varepsilon\, D_p\, \partial^2 C_p/\partial x^2$$

where ρ is the material's bulk density. For conditions where sorption is linear, the following equation can be written:

$$\partial C_p/\partial t = D_e/(\varepsilon + K_d\, \rho)\, \partial^2 C_p/\partial x^2 = D_a\, \partial^2 C_p/\partial x^2$$

where K_d is the distribution coefficient (see section 2.7.5), D_e is the is the effective and D_a the apparent diffusion constant, which are defined as follows:

$$D_e = \varepsilon D_p + K_d \rho\, D_s$$

$$D_a = D_e/(\varepsilon + K_d \rho)$$

When sorbing solutes are present at trace levels, sorption is independent of their concentration (i.e. where the concept of a linear sorption isotherm applies), C_p can be replaced by the total concentration in the porous material by multiplying by $\varepsilon + K_d \rho$ (the capacity factor, α) in the previous equation, which gives:

$$\partial C/\partial t = D_a\, \partial^2 C/\partial x^2$$

where C is the total concentration. This is the equation that is normally used for diffusion studies of radionuclides in bentonite. For diffusion of inert, non-interacting species, D_a is equal to D_e/ε (since anions interact with the negative charge of clay surfaces, this relation does not apply to anions).

Enhanced apparent cation diffusion: It has been observed in several cases that the apparent diffusion coefficient (D_a) for cations and their respective distribution coefficient (K_d) have not agreed as expected. D_a has been higher than expected from K_d and traditional porewater diffusion-sorption theory. While several explanations of this phenomenon have been proposed (see below), it is of foremost importance to recognise that an agreement of D_a and K_d is only to be expected for self-consistent datasets. I.e. both D_a and K_d need to correspond to the same set of conditions, in particular to the same porewater composition. Thus, several of the discrepancies between D_a and K_d observed in the past can be traced to the fact that incompatible conditions have been compared. In particular, D_a values that invariably correspond to the pore water composition in compacted bentonite have been compared directly with K_d values from batch experiments involving typically much more dilute solutions. Given that K_d values are derived for relevant porewater conditions, sorption and apparent diffusion coefficients for many elements agree within the overall data uncertainty /Ochs et al, 2003a; Bradbury and Baeyens, 2003/.

Nevertheless, there are cases where it seems clear that certain cations may have enhanced diffusivities, which must be taken into consideration in safety assessments. The presently available information suggests that these cases are largely restricted to mobile and cationic elements: Cs, and sometimes Sr and Ra /Ochs et al, 2003a/.

Enhanced apparent cation diffusion can be explained by a cation excess in the diffuse layer (the outer portion of the electrical double layer, EDL) extending from negatively charged clay surfaces into the pore space /Kato et al, 1995; Sato et al, 1995; Ochs et al, 2001/, see also section 2.7.5. Note that cation excess (as well as anion deficit, see below) have been well established in clay chemistry /see e.g. van Olphen, 1991/. Cations thus have an alternative transport pathway where their dissolved concentrations, and therefore the respective concentration gradients, are higher than in the actual pore solution (even though the diffusion coefficient in the diffuse layer could be lower than in the pore solution). Note that this additional pathway is available to cations that are accumulated in the diffuse layer, thereby retaining their mobility parallel to (but not perpendicular to) a mineral surface; it is not available to specifically (i.e. chemically) sorbed surface species formed at surface complexation or ion exchange sites.

While presumably based on the same underlying process, the so-called surface diffusion mechanism /Muurinen, 1994; Eriksen and Jansson, 1996; Yu and Neretnieks, 1997/ makes a direct link with actual sorption by introducing an additional surface diffusion coefficient that is multiplied with K_d. As pointed out above, however, there is no reason to assume that specifically sorbed species retain their mobility. A reduction of K_d in highly-compacted bentonite due to a decreased accessibility of surface sites had also been considered as a possible explanation for the disagreement of diffusion data with batch K_d values /Wanner et al/, but it was shown by /Kato et al, 1995/ that the entire physical porosity in compacted bentonite is acessible.

Anion exclusion: With increasing density, the porewidth in compacted bentonite decreases. At high degrees of compaction, the porewidth is small enough to cause a superimposition of the electrical double layers between two negatively charged pore walls. At this point, the anion deficit in the diffuse layer pointed out above significantly decreases the available diffusion pathway for anions, giving rise to a phenomenon known as anion exclusion. Evidence from several different studies suggests that the overlap of EDLs occurs at a dry density of about 1200–1500 kg/m^3 /Kozaki et al, 1998; Karnland, 1997a; Ochs et al, 2001/. The effect of anion exclusion becomes less at high salinities, because high ionic strength leads to a depression of the EDL /Stumm and Morgan, 1996/, and in sand-bentonite mixtures it is negligible. The safety-related importance of anion exclusion is considerable – it is one of the few phenomena that limit the transport of non-sorbing anions from a damaged canister.

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature		probably	possibly through thermal expansion, influence on EDL properties (no clear data available)
Water content	No	(Yes)	assumed saturated conditions
Gas content	No	(Yes)	assumed saturated conditions
Hydrovariables (pressure and flows)	No	(Yes)	assumed saturated conditions
Buffer geometry		Yes	through distribution of pressure??
Pore geometry	No	Yes	
Stress state	No	No	
Montmorillonite composition	No	Yes	extend of EDL depends on magnitude of charge and through physical porewidth
Pore water composition	possibly?	Yes	extend of EDL depends on ionic strength
Bentonite composition	No	Yes	through influence on porewater composition
Structural and stray materials	No	Yes	through influence on physical porewidth

Influence of sorption: The influence of sorption is taken into account in the equation for D_a.

Influence of buffer density: The transport of radionuclides in bentonite is influenced by the density of the material (the degree of compaction). This is due to a variation in porosity, as well as altered sorption in cases where soluble impurities lead to an increase of ionic strength as a function of density. In most experiments, the apparent diffusivity decreases with increasing density.

Influnce of montmorillonite composition: The diffusion properties are dependent on the interlamellar space in the bentonite which is in turn dependent on the permanent charge characteristics and the composition of the exchangeable cations.

Influence of speciation: Different species of the same element can have radically different diffusivities. This holds for free water diffusivities, but in particular for diffusion in bentonite pores. Due to their interaction with the diffuse layer extending from negative clay surfaces, diffusivities decrease in the order anionic species < neutral species < cationic species. For example, strontium has lower diffusivity at high concentration of sulphate, which can be explained by the formation of the neutral complex $SrSO_4$ /Ochs et al, 2001/.

Boundary conditions

The transfer of species from the bentonite to the rock can be visualised as the diffusion of dissolved species from the bentonite into flowing water in the near-field fracture network (equivalent flow rate or Qeq). The equivalent flow rate can be derived by solving the boundary layer theory equations for diffusive transport into flowing water. The Qeq depends on the geometry of the contact area, the water flux, the flow porosity and the water diffusivity.

Model studies/experimental studies

A large number of investigations are described in the literature concerning diffusion experiments in bentonite and similar clays; see /Yu and Neretnieks, 1997/ for a good overview and compilation of diffusion coefficients. Additional results form more recent studies are discussed in the data report /Ochs and Talerico, 2004/. Due to experimental reasons, most diffusion studies are performed as transient in-diffusion experiments. These experiments yield D_a, which is a lump-sum representation of all processes relevant for radionuclide migration, including diffusion and sorption. This holds in particular for moderately and strongly sorbing radionuclides. Steady-state through-diffusion studies, which can yield D_e and porosity, are largely restricted to mobile tracers (Cs, HTO, anions).

/Yu and Neretnieks, 1997/ also give a detailed discussion of different experimental methods and how the results can be interpreted. For comparisons of data from different studies, it is important to observe which technique has been used and how the diffusivities have been determined. If, for example, metal filters have been used, they can greatly influence the results. It also has to be realised in this respect that the extraction of diffusion coefficients invariably involves fitting of experimental data to the diffusion equations described above.

Few attempts have been made so far to derive diffusion coefficients through semi-mechanistic models. Some examples are discussed above in the context of anion exclusion and enhanced cation diffusion.

Natural analogues/observations in nature

The usefulness of natural analogues for determining diffusion constants is limited due to the difficulty of determining what conditions existed in the past.

Time perspective

Diffusion processes in the buffer material are of the greatest importance on all timescales. If the canister is intact, the process is of importance for the stability of the canister and the buffer. In the event of a defective canister, slow diffusion is very important in delaying, reducing and in many cases completely preventing releases of radionuclides.

Handling in the safety assessment SR-Can

Before saturation: The process is neglected since advection dominates.

After saturation: The process is included in the scenario specific modelling of the buffer chemical evolution for the thermal phase and the post-thermal long-term phase, see heading "Handling" for process 2.7.6. The process is treated in a simplified way with identical diffusivity for all elements in the buffer chemical evolution.

Failed canister: Included in the modelling of radionuclide transport for the long-term phase, see heading "Handling" for process 2.8.1. For the treatment of radionuclide diffusion, element specific diffusivities are used.

Boundary conditions: See sections referred to above.

Influences and couplings: Dependence on density, expected ionic charge of diffusing species, ionic strength and temperature will be considered in selecting diffusion constants for transport of radionuclides. The treatment of the buffer chemical evolution is currently less developed in this respect.

Uncertainties

Uncertainties in mechanistic understanding

The basic processes of enhanced cation diffusion and anion exclusion have been discussed above. Because of the experimental inaccessibility of bentonite pores, diffusion data are of macroscopic nature and are not well suited for any mechanistic interpretation. On a mechanistic level, the main shortcomings probably lie in the understanding of EDL properties as well as porewater structure and their influence on diffusion (and sorption) processes in compacted bentonite. This is particularly true for high densities that lead to an overlap of opposing EDLs.

The processes giving rise to the phenomena of enhanced cation diffusion and anion exclusion are the establishment of a cation excess and an anion deficit in the diffuse layer extending from a negatively charged mineral surface. Thus, these phenomena are caused by effects of enhanced or diminished ion concentrations in the volume of the porespace, rather than actual available porosity or different diffusivities. However, several approaches can be used to include the effects of this process in the diffusion equation that is used to define D_a: /Sato et al, 1995; Ochs et al, 2001/ expressed both cation excess and anion deficit through a concentration-correction factor for constrictivity (which leads to a decrease of D_e for anions and increase of D_e for cations), while assuming that the entire physical porosity is available for diffusion. On the other hand, it may be more practical to express this correction factor directly through the diffusion-available porosity, as the anion deficit in the diffuse layer leads to a decrease of the capacity factor α (which includes porosity, but not constrictivity). In clay chemistry, anion exclusion is also expressed by actual negative sorption /van Olphen, 1991/, which would also decrease α. Similarly, cation excess and sorption will increase α.

For the selection of radionuclide diffusion data and uncertainties for SR-Can /Ochs and Talerico, 2004/, anion exclusion was taken into account through a decrease in D_e and a reduced diffusion-available porosity. Enhanced cation diffusion was accounted for by an increase in D_e in combination with the physical porosity.

Model simplification uncertainties for the above handling in SR-Can

As pointed out above, diffusion is represented in a simplified way, through the use of selected constant effective diffusion coefficients and available porosities in the relevant transport codes (see sections 2.7.6 and 2.8.1). In that sense, the diffusion input parameters have to be viewed as conditional; i.e. their application will only be valid under the conditions considered in data derivation. Therefore, uncertainties will be related to the selection of consistent conditions. If, for example, the THM processes occurring during buffer evolution will lead to a significant alteration of bentonite properties, it may be necessary to use input parameters that differ form those for standard MX-80 bentonite.

In principle, this situation could be resolved by directly including diffusion processes in the model used for consequence calculations, which would require a THMC model approach. However, in addition to the great difficulties still associated with coupling THM processes on the one and C processes on the other hand, the present state of the art of quantifying radionuclide diffusion in compacted bentonite does not allow this. To date, semi-mechanistic models have been proposed and tested only for a few elements (Cs, Sr, anions; see above). At the same time, in case of many relevant radionuclides there is a clear lack of systematic data from through-diffusion experiments that would be needed to develop further diffusion models in a meaningful way.

While the abovementioned models are clearly still simplified, they appear to be quite robust as shown by the results of blind model predictions that are consistent with experimental data for several bentonites and over a range of densities /Ochs et al, 2001/. The respective model outputs are also consistent with the selections made for SR-Can. In addition, /Ochs and Talerico, 2004/ used the selected D_e values and corresponding K_d values for each element (and relevant oxidation state) to calculate apparent diffusivities, which were then compared to independent experimental data to provide an additional evaluation of the consistency of selected sorption and diffusion parameters. Based on the present evidence, it is expected that the direct use of diffusion parameters will not introduce significant additional uncertainties, given that important boundary conditions in SR-Can will be similar to the conditions considered in the derivation of input data.

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

Input data to SR-Can are effective diffusivities and available porosities to be directly used in consequence calculations. Data and uncertainties as a function of bentonite density are given in /Ochs and Talerico, 2004/, from which data for consequence calculations are chosen in the SR-Can data report.

2.5.4 Colloid transport

Overview/general description

Particles with sizes of the order of 10^{-9} to 10^{-6} m (colloids) could form on dissolution of the fuel. The diffusive transport of fuel colloids through highly-compacted bentonite is, however, assumed to be negligible, due to the tortuosity and small size of the bentonite pores. At emplacement the bentonite has a water ratio of 17%, which corresponds to a water film with a thickness of $6 \cdot 10^{-10}$ m between the individual montmorillonite flakes (inter-lamellar distance). Larger pores, between the original bentonite grains from which the blocks are produced, make up the remaining pore space. The water film thickness increases during water saturation, and the final theoretical mean inter-lamellar distance is around 10^{-9} m, which obviously would dramatically reduce colloid transport. However, a complete homogenization of the bentonite can not be expected, and a certain variation in the final pore size will remain also at full saturation. The disjoining force (swelling pressure), and internal friction which counteract the homogenization may be seen as governing variables.

Experiments with 15 nm gold colloids show that the microstructure of a bentonite with a dry density of 1,000 kg/m^3 effectively filters gold colloids /Kurosawa et al, 1997/. This corresponds to a saturated clay density of about 1640 kg/m^3. However, organic colloids will diffuse through bentonite, but the transport capacity is limited.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	No?	
Water content	No	(Yes?)	
Gas content	No	No	
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	No	No	
Pore geometry	No	Yes	
Stress stare	No	No	
Montmorillonite composition	No	Yes	
Pore water composition	No	Yes	
Bentonite composition	No	Yes	
Structural and stray materials	No	No	

Boundary conditions

The process is only interesting for the transport of colloids from the canister interior to the rock.

Model studies/experimental studies

/Kurosawa et al, 1997/ reports that 15 nm gold colloids are effectively filtered out at a dry density of 1000 kg/m^3 for bentonite/sand mixtures with a sand content of up to 40%. At 50% sand there is an indication of colloid breakthrough, but even there the retention is strong. No colloid breakthrough is observed for a 50/50 mixture when the dry density is increased to 1800 kg/m^3.

/Wold, 2003/ resports about diffusion experiments with organic colloids through MX-80 bentonite. The lignosulfonate (LS) and humic acid (HA) colloids used had an average size of 80 and <10 nm respectively, but the size distribution was broad. The LS and HA colloids showed diffusivities in the same range as negatively charged ions like Cl^- and I^-, which is surprising, especially for LS, since they also are negatively charged and the intra-lammelar spacing in a saturated bentonite is ~15 Å. One possible explanation for lack of colloid filtration could be the broad size distribution of the LS colloid. A size distribution change could not be detected due to the experimental conditions.

Time perspective

The process is important for all repository timescales, but only if canisters have failed.

Natural analogues/observations in nature

Handling in the safety assessment SR-Can

The concern in the safety assessment is the transport of radionuclides with colloids from a potentially failed canister, in particular fuel colloids. This transport mechanism will be neglected, provided the buffer completely envelops the canister and has a dry density of at least 1,000 kg/m^3, i.e. a clay density of at least 1650 kg/m^3.

The effect of a loss of the colloid filtration effect will potentially be treated as a residual scenario. Transport of radionuclides in/sorbed on a colloid phase would mean that the concept of solubility limits is not valid. Radionuclide solubilties would have to be adjusted for such a scenario.

Uncertainties

Uncertainties in mechanistic understanding

As seen in the Model/Experiment section above, different types of colloids behave very differently. It is not clear why there is such a big difference between inorganic and organic colloids. This issue needs to be addressed by additional experimental work.

Model simplification uncertainties for the above handling in SR-Can

No model is used.

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

No data are used.

2.5.5 Sorption (including ion exchange)

Overview/general description

Radionuclides and major ions in the buffer's pore water can be bound to the surfaces of the bentonite material in several ways (see below). Together, these processes are termed sorption and are of essential importance for the function of the buffer, since they drastically affect the mobility of most radionuclides, but also of major ions which is in turn related to the evolution of the buffer. The principal mineral in bentonite is montmorillonite, which consists of octahedral alumina sheets sandwiched between tetrahedral silica sheets (2:1 clay). Such clay minerals feature two distinctly different types of surfaces, where two main types of sorption take place /e.g. Sposito, 1984; Stumm and Morgan, 1996/.

- The siloxane ('layer') surfaces of clay minerals are *permanent charge surfaces.* These charges can derive from isomorphous substitutions, which result in a constant negative surface charge. A macroscopically electrically neutral surface involves charge-compensating cations, and sorption takes place when compensating ions are exchanged. *Ion exchange models* have been used in soil science since the first quarter of the 20th century. Several formalisms have been developed to correct for activity changes of the exchanger as a function of the composition of the charge-compensating ions. The most common are the equivalent fraction (Gaines-Thomas, GT) and mole fraction (Vanselow) models.
- The edge surfaces of clay minerals are *variable charge surfaces*. They carry a net positive or negative surface charge depending on the species sorbed to their surfaces (potential-determining ions), often involving surface-bound OH^- groups. *Surface complexation and ligand exhange models* were established by Stumm, Schindler and co-workers in the 1970s by extending proton-binding and metal coordination chemistry in a rigorous fashion to surface chemistry. To account for the electrostatic field, the mass laws for surface equilibria often include an electrostatic correction term. The scientific basis for these corrections is derived from Electrical Double Layer (EDL) theory.

Ion exchange is the typical sorption mechanism for alkali, and alkaline-earth elements, as well as transition metals at low pH values where positive species are predominant. Surface complexation is generally the more relevant process for all reactive elements (transition metals, actinides, lanthanides, reactive anions such as carbonate). Note that both ion exchange and surface complexation take place simultaneously, but at different surfaces.

The processes can be roughly divided into ion exchange and alteration of the total quantity of cations. The most important factor for both processes is the ion content of the pore water, which is principally determined by the original cations in the montmorillonite's exchange positions, readily soluble minerals in the original bentonite material, and by the surrounding groundwater.

H^+ ions are bound to the edge sites of clays (as well as to ion exchange positions at very low pH), which means that pH changes can occur due to surface chemical reactions. Due to its ampholytic properties, i.e. ability to both receive and donate hydrogen ions, the montmorillonite can counteract external changes in pH /Wanner et al/.

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

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	Yes	influence of temperature on sorption must be acknowledged, but effect is not clear
Water content	No	No	
Gas content	No	No	
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	No	No	
Pore geometry	Yes	Yes	through influence on EDL properties
Stress state	Yes	Yes	through influence on both porewater composition and EDL properties
Montmorillonite composition	Yes	Yes (surface composition)	
Pore water composition	Yes	Yes	
Bentonite composition	Yes	Yes	
Structural and stray materials	Yes	Yes	

Influence on pore geometry, hydraulic conductivity and swelling pressure in the buffer: The volume of the buffer is so constrained that a maximum swelling of the bentonite cannot be achieved even after complete conversion to the Ca-form. The pore geometry, and thereby also the hydraulic conductivity, are therefore not expected to be significantly altered by this process. Very high ion concentration in the pore water leads to a significant reduction in the swelling pressure due to osmotic effects and a suppresion of the EDL, and to an increase in the hydraulic conductivity (see section 2.6.1). A maximum content of sodium chloride (36 percent, equivalent to saturated solution) leads to a reduction in the swelling pressure by approximately 50 percent, and to an increase in the hydraulic conductivity by approximately a factor of 10 (see also section 2.7.8).

Influence of density: Under a given set of conditions, sorption increases if the amount of available surface area (i.e. the solid/water ratio) is increased. By expressing sorption through K_d (see below), which includes the solid/water ratio, this effect is already taken into account. Given that the solution composition remains constant, K_d is clearly independent of the solid/water ratio and, therefore, of the buffer density. It appears from diffusion experiments that the apparent K_d is influenced by density, for example the K_d for caesium is halved when the density of the water-saturated system changes from 1,300 to 1,950 kg/m^3. However, this can be entirely attributed to an increase of ionic strength caused by the dissolution of impurities from an increasing mass of bentonite, and is easily reproduced by simple thermodynamic sorption models /Ochs et al, 2001/. This effect is mostly relevant in case of dilute groundwaters.

Influence of temperature: Most experiments have been carried out at room temperature, and not enough sorption data as a function of temperature are available to clearly evaluate temperature effects. In general, a decrease of sorption with temperature would be expected, but some isolated studies /e.g. Lu et al, 2003/ indicate the opposite. Thus, an increase or decrease of the temperature is potentially of importance for sorption, but the effects within the temperature range expected in the repository are deemed to be covered with good margin by other uncertainties.

Boundary conditions

There are no particular boundary conditions to discuss for this process. The relevant boundary conditions in order to treat the process quantitatively are those of the transport processes that control the exchange of solutes between the buffer pore water and the water in its adjacent components, i.e. the boundary conditions of the processes diffusion and advection.

Model studies/experimental studies

Ion exchange as well as surface complexation are well-established process and quantitative thermodynamic models exist for calculation of equilibrium states. The relation of these sorption processes to the different surfaces available on montmorillonite has been discussed above.

Effect of sorption on buffer properties: Conversion from sodium to calcium states by ion exchange has been modelled for deep repository conditions (taking into account all other ion exchange as well as edge site protolysis equilibria). Calculations have been carried out for expected repository conditions with respect to the content of accessory minerals (calcite, gypsum, halide) and groundwater flows /Wanner et al, 1992 /. See also the processes 2.5.2 and 2.7.8.

Effect of sorption on radionuclide transport: In assessing radionuclide transport out from a repository, the sorption equilibrium is normally described as a linear relationship between the sorbed concentration and the concentration in solution, characterized by a distribution coefficient K_d , which is defined as

$$K_d = q / C_w = K_d = \left(\frac{C_{init} - C_{equil}}{C_{equil}} \right) \frac{v}{m}$$

where:

C_{init} is the initial aqueous concentration of a key element [mol/m^3] as measured in the reference solutions without solids

C_{equil} is the final equilibrium aqueous concentration of this element [mol/m^3]

C_w is the concentration in solution

v is the volume of the solution used for a batch experiment [m^3]

m is the mass of the solid phase used for a batch experiment [kg]

q is the sorbed concentration as mass per weight unit of solid phase

When the concentration of the species in solution is low (more specifically, at a state sufficiently far from surface saturation), which is normally the case for radionuclides, the linear approximation is justified. K_d can also be calculated in a straightforward fashion from the output of thermodynamic sorption models:

$$K_d \,[\text{m}^3/\text{kg}] = \frac{\text{stoichiometric sum of surface species of element X [mol/kg]}}{\text{stoichiometric sum of solution species of element X [mol/m}^3\text{]}}$$

Surface complexation models with parameters from well-controlled experiments that include sufficient variations of geochemical key parameters can often be used to describe the sorption's dependence on external parameters such as pH, etc, see e.g. examples in /NEA, 2001/.

Time perspective

An ion exchange process in itself is fast, so the dissolution rate of other minerals and the transport rate in groundwater and bentonite pores will control the ion exchange from Na^+ to Ca^{2+}. Relevant changes in buffer properties are expected to occur on the hundred-thousand-year scale /Bruno et al, 1999/. It has to be pointed out further that modelling the conversion of Na- to Ca-bentonite by implementing ion exchange in a mixing-tank type of model, as in /Bruno et al, 1999/, is in all likelihood a pessimistic approach, in the sense that the it overestimates the rate of the exchange process: To compensate the diffuse layer charge in the porespace, a certain cation concentration is required /Ochs and Talerico, 2003b/. While the maximum concentration of Ca is limited by the solubility of calcite or gypsum, the concentration of Na is not limited, and it can be doubted that a full conversion to the Ca-form is possible under these conditions.

Sorption in the buffer is an important process for the retention of many radionuclides. The timescales that are of interest are dependent on the half-life of the individual nuclide. If the travel time through the buffer is of the same order of magnitude or longer, sorption is of great importance for the amount released. If the travel time through the buffer, including retardation from sorption, is short in comparison to half-life, the sorption process is less important.

Natural analogues/observations in nature

Many bentonite deposits contain pore water with high salinities. Investigations of effects of high salinity in natural smectites exist but have not been utilized specifically for repository-related questions. Natural analogues also offer evidence that migration of sorbing radionuclides occurred only over limited distances away from a source in clay-rich substrates /Smellie and Karlsson, 1996/

Handling in the safety assessment SR-Can

Before saturation: The process is neglected.

After saturation: Ion exchange is included in the scenario specific modelling of the buffer chemical evolution for the thermal phase and the post-thermal long-term phase, see heading "Handling" for process 2.7.6.

Failed canister: Sorption is included in the modelling of radionuclide transport for the long-term phase, see heading "Handling" for process 2.8.1.

Boundary conditions: See sections referred to above.

Influences and couplings: Dependence on composition and pH of the porewater in the buffer, as resulting from buffer properties and groundwater composition, will be considered in selecting sorption constants for radionuclide transport. Because of the conditional nature of sorption constants, the data selected need to be derived for the relevant safety assessment conditions.

Uncertainties

Uncertainties in mechanistic understanding

Both conceptual understanding and a large quantity of measurement data exist for simplified systems.. Detailed reviews of sorption processes, underlying experimental data, model results, as well as the relation to performance assessment can be found in e.g. /NEA, 2001; Davis and Kent, 1990/. In particular, it has been shown that thermodynamic sorption models developed for purified Na-montmorillonite can be applied to Ca-montmorillonite /Bradbury and Baeyens, 1999/, and that such models give results for compacted bentonite that are compatible with diffusion data /Ochs et al, 2003a/. To date, there is no doubt that sorption processes are well understood in a quantitative way and in terms of thermodynamic description, and that this understanding can be used to make educated predictions of K_d values to be used in safety assessment calculations. However, there is a clear lack of data for understanding and predicting the influence of some important variables (such as dissolved carbonate concentration) on radionuclide sorption, and this has to be taken into account in evaluating uncertainties of K_d values /Ochs and Talerico, 2004/. In case of some radionuclides, the underlying thermodynamic database is insufficient to quantitatively evaluate their behaviour for different conditions.

Model simplification uncertainties for the above handling in SR-Can

As pointed out above, the K_d concept for radionuclide transport modelling is basically justified under conditions far from surface saturation, which can be expected in the case of diffusion-limited radionuclide migration in a bentonite buffer. At the same time, it is clear that a K_d is a highly conditional parameter in terms of chemical conditions (pH, ionic strength, etc) and has to be derived for each set of conditions. This was done in case of the data selection for SR-Can using a semi-quantitative, traceable procedure to transfer K_d data from experimental to safety assessment-specific conditions /Ochs and Talerico, 2004/. For several elements, it was possible to use state of the art thermodynamic sorption models to derive K_d values for the specified conditions. The model-derived data were in all cases within the uncertainty range of the values that had been derived using the semi-quantitative conversion procedure. This strongly suggests that for the specified conditions, direct use of a sorption model and use of recommended K_d values would lead to consistent results. To avoid potential problems related to the direct use of K_d in cases where conditions may vary to some degree (or to avoid re-derivation for each condition), thermodynamic sorption models could be directly coupled with transport codes (see section 2.7.3). Again, for many radionuclides not enough systematic sorption data for the relevant conditions are available to date to develop or sufficiently constrain thermodynamic models.

As long as the bentonite buffer can be expected to represent a homogeneous geochemical compartment, no spatial variability needs to be taken into account.

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

Input data to SR-Can are K_d values to be directly used in consequence calculations. K_d values for radionuclides for various conditions, as well as the associated uncertainties are given in /Ochs and Talerico, 2004/, from which data for consequence calculations are chosen in the SR-Can data report.

2.5.6 Alterations of impurities

Overview/general description

The buffer material consists not only of montmorillonite, but also other accessory minerals as well as impurities. In the repository environment, these can be dissolved and sometimes re-precipitated depending on the prevailing conditions.

Precipitation of carbonates and sulphates in the areas nearest the canisters may lead to the formation of a porous contact zone between the buffer and the canister surface and an increase in the strength of the buffer. When the heat period is over and temperature gradients no longer exist in the buffer, it is likely that the precipitated solids will be dissolved to some degree and diffuse in ionic form out through the buffer. Precipitation of silicon may reduce the swelling potential, among other additional effects.

The composition of MX-80 and Deponit CA-N can be found in Table 2-1.

The carbonate and pyrite contents are of decisive importance for pH, Eh and alkalinity in the near field. Bentonite also contains small quantities of gypsum ($CaSO_4 \cdot 2H_2O$), which may be of importance in an early phase during the high temperature transient.

Evaporation of incoming ground-water during the saturation phase in the warmer parts may lead to different concentration of dissolved elements at different positions in the buffer.

The accessory minerals in bentonite are either stable or metastable in the environment where they were originally mined. In the repository, however, they will be exposed to a fluid which differs in some respects from that on the original site, in terms of both composition and temperature. Most accessory minerals in the bentonite are nevertheless stable in the granitic groundwaters prevailing in the Fenoscandian shield. There are, however, some dissolution-precipitation processes that are important:

Calcite dissolution: Calcite is stable in groundwater. In the bentonite, however, the ion exchange process will compete for the free calcium ions. The reaction

$2XNa + Ca^{2+} \leftrightarrow X_2Ca + 2Na^+$

competes with

$Ca^{2+} + HCO_3^- \leftrightarrow CaCO_3(s) + H^+$

Table 2-1. Impurities and accessory minerals in MX-80 and Deponit CA-N bentonite /Karnland et al, 2004/.

Component	MX-80 (wt-%)	Deponit CA-N (wt-%)	Uncertainty (± wt-%)
Calcite + Siderite	0	10	1
Quartz	3	1	0.5
Cristobalite	2	1	0.5
Pyrite	0.07	0.5	0.05
Mica	4	0	1
Gypsum	0.7	1.8 (anhydrite)	0.2
Albite	3	0	1
Dolomite	0	3	1
Montmorillonite	87	81	3
Na-	72%	24%	5
Ca-	18%	46%	5
Mg-	8%	29%	5
K-	2%	2%	1
Anorthoclase	0	2	1
CEC (meq/100g)	75	70	2

The carbonate contents of the groundwaters that may be encountered are sufficiently low for the ion exchange process to dominate and calcite will be dissolved. This will entail a net proton consumption, i.e. an increase in pH. The water flux in the buffer is very low, and the reactions are close to equilibrium.

Siderite dissolution and iron hydroxide precipitation: The presence of pure siderite or ankerite, a mixed calcium-iron carbonate is of paramount importance for the redox control of the system through the following reaction

$FeCO_3(s) + \frac{1}{4}O_2(aq) + 2\frac{1}{2}H_2O \leftrightarrow Fe(OH)_3(s) + HCO_3^- + H^+$

Siderite oxidation leads to an increase of iron in solution allowing the precipitation of iron (III) hydroxides and thus controlling the redox of the system. However, during the early stage, when high temperatures are expected in the buffer, hematite would be the stable iron (III) phase instead of iron (III) hydroxide, leading to the precipitation of hematite close to the canister and increasing the strength of the buffer.

Pyrite oxidation/dissolution: Pyrite is also stable in groundwater. However, penetrating oxygenated water can oxidise pyrite in accordance with

$2\ FeS_2(s) + 7\frac{1}{2}O_2(aq) + 7H_2O \leftrightarrow 2Fe(OH)_3(s) + 4SO_4^{2-} + 8H^+$

Pyrite oxidation releases protons and can thereby lower the pH, but this is buffered by the dissolution of calcite previously discussed. There is enough pyrite in the buffer (the MX-80 material) to consume all initial oxygen left after closure of the repository. The quantity is also sufficient to prevent any penetrating oxygenated water from coming into contact with the canister for hundreds of thousands of years. However, as this process is kinetically driven, it is likely that pyrite oxidation exerts a minor effect on the redox of the system, which will be controlled by the iron carbonate dissolution process.

Pyrite is a source of a sulphides that could act as a corrodant on the copper canister. Under educing conditions pyrite dissolves as:

$FeS_2(s)+H^+ \leftrightarrow Fe^{2+} + HS^- + S^0$

The concentration of HS^- is an important parameter for the canister corrosion.

Dissolution of calcium sulphates: Calcium sulphates (gypsum and anhydrite) and calcite have lower solubility at high temperatures. At an early stage, when the canister temperature is high, it is possible that they will be dissolved in the colder portion of the buffer and precipitate on the canister surface.

Precipitation/dissolution of silicon: The largest portion of the impurities in the bentonite consists of quartz and feldspars. Feldspars are not stable in the repository environment, although the kinetically driven dissolution is a relatively slow process which is accompanied by the precipitation (or replacement) of clay minerals, i.e. kaolinite.
The low water flux in the buffer is likely to minimise the kinetic dissolution of feldspars. Quartz is normally stable in the natural repository environment, but its solubility increases with increasing temperature. Silica will dissolve due to high temperature close the canister and will be transported by diffussion outwards into the colder parts where precipitation may take place. In the Buffer Mass Test in Stripa, the buffer was analyzed with respect to the distribution of silicon, but no definite conclusion could be drawn regarding possible enrichment in the coldest part /Pusch, 1985b/.

Most of the conceivable processes are in themselves well known and can be modelled for less complex systems. However, conditions in the buffer as far as transport and reaction kinetics are concerned are not fully understood for all processes. Transport of solutes can take place in different forms and cannot be fully described at present. In particular, modelling problems remain during the water saturation phase, when water is transported both in vapour and liquid form. Modelling of the system must therefore be coupled to the THM processes that proceed in parallel with transport and reaction kinetics.

Silicon is the most common element in the buffer and occurs in a number of different minerals and phases. The following processes are not fully understood:

- release of silicon from the different minerals,
- transport of dissolved silicon driven by the prevailing temperature gradient,
- precipitation of silicon minerals.

Precipitation of silicon is particularly complex, since it is dependent on several interacting factors and since several conceivable forms occur, both crystalline and amorphous.

The greatest uncertainty concerns the scope of cementation processes as a consequence of dissolution, transport and precipitation of silicon or aluminosilicate minerals. The scope and consequences of cementation cannot be predicted with reasonable certainty today.

The following table summarises how the process influences and is influenced by all buffer variables

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	Only very high radiation fields could influence the processes.
Temperature	No	Yes	Temperature highly influences the extent of precipitation/dissolution of secondary minerals.
Water content	Yes	Yes	Precipitation-dissolution of accessory minerals modifies the porosity.
Gas content	Yes	Yes	A secondary effect from water content variations.
Hydrovariables (pressure and flows)	Yes	Yes	Precipitation-dissolution of minerals
Buffer geometry	No	Yes	
Pore geometry	Yes	Yes	Precipitation-dissolution of minerals
Stress state	Yes	Yes	
Montmorillonite composition	Yes	Yes	
Pore water composition	Yes	Yes	
Bentonite composition	Yes	Yes	
Structural and stray materials	Yes	Yes	Seconday mineral formation may influence the alteration of structural components. This is particularly true for sulphates.

Boundary conditions

There are no particular boundary conditions to discuss for these processes. The relevant boundary conditions in order to treat the process quantitatively are those of the transport processes that control the exchange of solutes between the buffer pore water and the water in its adjacent components, i.e. the boundary conditions of the processes diffusion and advection.

For alterations of impurities, the exchange of a number of solutes like sodium and calcium ions, carbonate and oxygen is relevant.

Model studies/experimental studies

Hydrothermal tests with purified standard bentonite (SWY-1) heated to 150–200°C have shown that cooling leads to precipitation of silicon compounds in various forms. The precipitation is assumed to cause cementation effects, including a strength increase, which has been demonstrated in several laboratory investigations /Pusch and Karnland, 1988; Pusch et al, 1991/. According to these investigations, the scope of the precipitations is dependent on the temperature. At the highest temperature in the buffer (90°C), the cementation was not of such a great extent that it can be regarded as problematic. The tests were carried out without a temperature gradient, which cannot be regarded as pessimistic conditions.

Precipitation of sulphate and carbonate could be observed in one-year tests with hydrothermal treatment of MX-80. XRD analyses indicated that sulphates and calcite had gone into solution and been transported to the hot iron surface, where they had been precipitated. It is also probable that feldspars went into solution. Quartz was enriched at the colder boundary These are experimental evidences (as indicated in the first sentence), whereas the earlier statement refers to the quantification of this process through a geochemical model /Pusch et al, 1993/.

In the Buffer Mass Test in Stripa (max 90°C), the buffer was analyzed with respect to the distribution of silica, but no definite conclusion could be drawn regarding possible enrichment in the coldest part /Pusch, 1985b/.

In the LOT tests at Äspö HRL, buffer material is exposed to high temperature gradients (maximum130 to 80°C over a 10 cm distance). Material from the one year tests was analyses at 25 positions with respect to i.a. element distribution and rheological properties. No significant gradients were found for silica. The tensile strength was slightly reduced and the tensile strain was slightly increased, which shows that no cementation had taken place in the major part of the buffer material. However, precipitation of mainly gypsum was found in a 1–2 mm thick rim around the copper tube. The ongoing five-years tests are expected to give information on whether the precipitation is an effect of evaporation of the inflowing water as a part of the saturation process, or if the process continues also after full water saturation.

Several laboratory investigations have been carried out for the purpose of studying the distribution of readily soluble ions in the buffer's pore space after water uptake against a temperature gradient. Effects of the clay's density, degree of saturation, testing time, open or closed conditions, and ion content and pressure in the ambient water solution have been investigated. The experimental results show that the enrichment of solutes is insignificant in specimens with high clay density, and that a high original water content and high ambient water pressure reduce the enrichment in specimens with lower density /Karnland, 1995/. Similar studies are currently being conducted in field tests in Äspö (the LOT project) /Karnland et al, 2000/, and in laboratory experiments under the auspices of ENRESA.

Several geochemical models have been developed in order to predict the long term behaviour of the buffer under repository conditions. Some of the models are based on a mixing tank concept /Wanner et al, 1992; Bruno et al, 1999/ where the thermal stage has not been considered. Other models are reactive transport based models where the thermal stage is considered /Arcos et al, 2000/. All these models suggest that cation exchange is the dominant process in the buffer resulting in a direct control on the calcite precipitation-dissolution, thus buffering the pH of the system. Also the effect protonation-deprotonation of montmorillonite edge sites can play a role on the pH buffering. However, earlier models /Bruno et al, 1999; Arcos et al, 2000/ indicate that calcite equilibrium is the main process buffering pH, and only if calcite is exhausted edge sites will be able to buffer pH in the system. The redox buffering capacity of the bentonite is somewhat controversial according to these models. Some of the models predict the redox buffering is due to pyrite oxidation and iron (III) hydroxide precipitation; whereas other models pointed to the iron carbonate (siderite) dissolution and iron (III) oxy-hydroxides as the redox buffering capacity of the system.

An additional model concerning the LOT experiment at Äspö /Arcos et al, 2003/ has been developed leading to results similar to those obtained in previous models. Unfortunately there are still no experimental data to compare with. Unfortunately, when comparing the modelling results with experimental data /Domènech et al, 2004/ some disagreements have been observed. It is likely that these differences could be due to the uncertainty associated to the initial conditions selected for the model. Actually, these initial conditions are being revised in the frame of the LOT project.

Natural analogues/observations in nature

Development of salt crusts in arid regions resembles in principle certain conditions during the water saturation phase. Direct SKB-related studies have not been conducted, but a large body of literature exists on the subject.

Silicon precipitation occurs at a large number of places where hydrothermal transformation of bentonite has taken place. Numerous scientific articles have been published, some directly linked to repository questions, see for example /Pusch et al, 1998/.

At present, SKB is involved in the study of a bentonite natural analogue in Spain, the BARRA project, under the coordination of ENRESA. Two processes are being studied in this natural system: 1) the changes in the bentonite induced by long time exposition to very high salinity environment, and 2) the thermal effect induced by a sub-volcanic intrusion on the bentonite properties The initial results from this study show that exposure to very high salinities does not alter the properties of the bentonite.

Time perspective

The chemical precipitation/dissolution processes are most important at an early stage when the temperature is high and oxygen remains in the repository tunnels. At later stages, most of the processes go towards equilibrium. The accessory minerals in the bentonite are, however, important also in time scales of hundreds of thousands of years if the chemistry in the repository should be disturbed for some reason; both pH and Eh are buffered effectively by the minerals that are common in different bentonite materials.

Handling in the safety assessment SR-Can

Before saturation: Geochemical processes will be the same both before and after saturation phases. This is just a matter of water flow and solutes transport.

After saturation: The process is included in the scenario specific modelling of the buffer chemical evolution for the thermal phase and the post-thermal long-term phase. This modelling includes, in addition to alterations of impurities, also the processes diffusion (section 2.7.3), ion exchange (section 2.7.5) and aqueous speciation and reactions (section 2.7.7). In formulating initial conditions, the initial intrusion of groundwater during buffer saturation should be considered if that has a relevant influence on the initial conditions. The main model will be a 2D reactive transport model based on the geochemical models of the LOT experiment /Arcos et al, 2003; Domènech et al, 2004/. The model handles advective flow in a fracture in granite intersecting the deposition hole, diffusive transport in the bentonite, dissolution-precipitation of main bentonite accessory minerals, cation exchange in the bentonite, protonation-deprotonation reactions in the smectite fraction and precipitation of favoured secondary phases in both bentonite and granitic fracture. Prior to application in SR-Can, the model will be validated by comparing modelling results with experimental data from the LOT test.

In addition, a simplified mixed tank model, embedded in the system evolution model /Hedin, 2004/ will be used for preliminary calculations and to study sensitivities to input data.

Failed canister: In the case of a failed canister, vast amounts of metallic iron, originating from the corroding cast iron canister insert, will be added to the geochemical system. This will be included in the model mentioned above, in a dedicated calculation case.

Boundary conditions: Main model: The diffusive transfer of species between the flowing water and the bentonite is explicitly modelled. The system evolution model uses the Qeq concept to handle the boundary conditions. This is sufficient to estimate a general time scale on which the evolution occurs.

Influences and couplings: The modelling is in itself a coupling of the four (groups of) processes mentioned above and is thus a coupled modelling of most of the significant long-term chemical processes. A large number of the variable influences related to the chemical conditions (pore water composition, montmorillonite composition, bentonite composition) are also included.

The changes in the buffer accessory mineral content as a consequence of the thermal stage has been modelled, but not validated through a comparison with experimental and/or natural analogue data. This makes it difficult to ensure that changes due to the thermal stage are minor or that these results should be assumed to be the initial conditions for the low temperature stage.

In addition to the processes discussed above, the behaviour of silica and clay minerals and their effect on cementation could be addressed by performing some scoping calculations considering the present knowledge on kinetic dissolution-precipitation of smectite, silica-phases and the precipitation of alternative clay minerals (kaolinite and/or illite). These scoping calculations would give some qualitative information on the relative importance of these processes and the effect on bentonite properties, i.e. porosity changes. See also section 2.7.9.

Uncertainties

Uncertainties in mechanistic understanding

Some critical uncertainties remain concerning the mechanistic understanding of the processes that control the redox state of the bentonite system. It is not clear yet to which extent pyrite and/or siderite are the main redox controlling phases, although either process, or their combination, will scavenge the remaining oxygen in the near field. This should be discerned by comparative modelling of pyrite and siderite kinetic availability.

In addition, most of the secondary precipitation/dissolution processes which are temperature driven are thermodynamically controlled. Hence, there is no need for a detailed mechanistic understanding. However, some of the silicate transformations are kinetically controlled and their mechanistic understanding in compacted bentonite conditions is poor. This is mainly related to clay mineral transformations (dissolution of smectite and precipitation of illite and/or kaolinite). Although these reactions occur at a very slow rate, their effect on the overall geochemical evolution is not yet clear.

Model simplification uncertainties for the above handling in SR-Can

Secondary dissolution and precipitation processes have not been fully implemented in a coupled model of buffer evolution. The processes are only considered when required for the definition of critical master variables in the system (pH and pe). A possibility could be to couple the THM processes occurring with the key chemical processes that will control the geochemical evolution of the buffer system.

There are a number of accessory minerals, such as silicates other than silica, which are not included in the models, as most of them tend to dissolve at a very slow kinetic rate. Other silicates (usually clay minerals) precipitate at similarly slow rates. However, the dissolution-precipitation processes involving these two groups of minerals could affect the stability of montmorillonite as well as the chemical composition of the buffer pore water, thus affecting the chemical long term evolution of the buffer. See also section 2.7.9.

There is no model accounting for the dissolution/precipitation of accessory minerals during the water saturation stage of the buffer. As water flux will be faster than when the buffer is water saturated and the buffer composition will be far from equilibrium with incoming water, some changes will occur faster. These changes will account for step compositional gradients in the buffer and changes in porosity as accessory minerals dissolve and/or precipitate, leading to changes in hydraulic properties of the buffer. Thus, modelling of the system must therefore be coupled to the THM processes that proceed in parallel with transport and chemical reactions. A submodel dealing with the saturation stage of the bentonite will, at least, give an idea on the magnitude of theses changes.

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

Key thermodynamic data are well established and therefore reliable. The main uncertainties remain in the definition of the kinetically controlled processes. These are linked to the mechanistic uncertainties previously mentioned.

Proper data for surface reactions (i.e. cation exchange and edge site reactions) is of paramount importance for the correct assessment of the bentonite behaviour. Two types of information are needed for geochemical modelling: i) Cation occupancy of the CEC and protonation state of the smectite surface, and ii) Exchange and protonation-deprotonation constants. The main uncertainty associated with these data is related to the experimental conditions used for their determination (i.e. low bentonite density and high solid/liquid ratios). Recent experiments have addressed this problem, however there is still a large uncertainty related to the determination of cation exchange constants and their variation as a function of bentonite density. These data and the associated uncertainties will tentatively be handled in the SR-Can data report.

2.5.7 Aqueous speciation and reactions

When the buffer is saturated, the water content is about 40%. The composition of this water will be determined by the composition of the pore water present at deposition, of the composition of groundwater on the site, reactions with the buffer and canister material and the impurities in the bentonite. Within the aqueous phase chemical reactions will take place and the final composition of the water can be determined with thermodynamics and kinetics. Examples of this are reactions like:

$HCO_3^- \leftrightarrow CO_3^{2-} + H^+$ and $HS^- + H^+ \leftrightarrow H_2S(aq)$

Overview/general description

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	Possibly	Radiolysis
Temperature	No	Yes	
Water content	No	No	
Gas content	No	No	
Hydrovariables (pressure and flows)	No	Yes	
Buffer geometry	No	No	
Pore geometry	No	No	
Stress state	Yes	Yes	See 2.7.8
Montmorillonite composition	Yes	Yes	See 2.7.9
Pore water composition	Yes	Yes	
Bentonite composition	No	No	See 2.7.9 and 2.7.6
Structural and stray materials	No	No	

Boundary conditions

By definition, this process takes place in the pore water. The boundary conditions will be the canister, the groundwater, the bentonite and the impurities.

Model studies/experimental studies

Direct measurements of the porewater composition in compacted bentonite is impossible. Limited amounts of water can be squeezed out under high pressure, but the composition of that water may not be representative of the porewater. The amount of non-sorbing elements with high solubility can be measured by dispersion of the bentonite in water, still this only gives the total amount and reveals nothing about the speciation in the bentonite.

Modelling of the chemical composition/speciation of the porewater is done with an integrated model for the chemical evolution in the near field. This is described in 2.7.6. Speciation of radionuclides is discussed in upcoming process report for the Fuel, See also the process report for SR 97 /SKB, 1999/.

Natural analogues/observations in nature

Not applicable.

Time perspective

The process is important for all repository time scales.

Handling in the safety assessment SR-Can

Before saturation: Geochemical processes will be the same before and after saturation. This is just a matter of water flow and solutes transport, something to be considered in other sections.

After saturation: The process is included in the scenario specific modelling of the buffer chemical evolution for the thermal phase and the post-thermal long-term phase. This is described in section 2.7.6

Failed canister: In the case of a failed canister, vast amounts of metallic iron, originating from the cast iron canister insert, will be added to the geochemical system. This will be included in the model mentioned above, as a dedicated calculation case.

Boundary conditions: See section 2.7.6.

Influences and couplings: The modelling is in itself a coupling of the four (groups of) processes mentioned above and is thus a coupled modelling of most of the significant long-term chemical processes. A large number of the variable influences related to the chemical conditions (pore water composition, montmorillonite composition, bentonite composition) are also included.

Uncertainties

Uncertainties in mechanistic understanding

The model assumes that the chemistry in porewater is identical to the chemistry in liquid water. The effect of the bentonite surface and structure is not included (except the ion-exchange). It is not clear if or how this will affect the chemistry. See also section 2.7.8.

Model simplification uncertainties for the above handling in SR-Can

See section 2.7.6

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

See section 2.7.6

2.5.8 Osmosis

Overview/general description

A classical view of osmosis, as shown in Figure 2-11, may be used in order to illustrate the conditions in a bentonite buffer. The two sides of the water system are divided by a semi-permeable membrane only permeable to water molecules. If salt is added to the left side, the water activity is reduced and a transport of water from right to left will take place. The left water table will thereby rise and a hydrostatic pressure develops (a). If salt is now added to the right side, there will be a leveling of the water tables on both sides (b). If the volume change, in the first place, is instead prohibited by force, a pressure will rise in the salt solution (c), corresponding to the previous hydrostatic pressure. Introduction of salt into the right hand side of the membrane will lead to a drop in pressure. A complete drop of pressure will evidently take place when the two concentrations are equaled (d).

The analog to bentonite buffer is that the left side of the system represents the bentonite and the right side an external ground-water solution. An external solution would reduce the pressure produced by the clay, to an extent equal to the osmotic pressure of the solution, if no ions could pass into the clay. A sodium chloride concentration of around 1.7 M (10% by weight) would then result in a complete loss of swelling pressure in a KBS-3 buffer. On the other hand, if ions could pass freely into the clay, then the final conditions would be equal concentrations on both sides and no effect on swelling pressure would be found, neither of which is in agreement with experimental results. Laboratory experiments show that ions from the external solution do pass into the clay to a certain extent, and the difference in concentration between external and introduced ions leads to a drop in the swelling pressure /Karnland, 1997/.

The conditions in the bentonite/ground-water system are typical for what is often referred to as a Donnan equilibrium system /Donnan, 1911/. The equilibrium is characterized by one ion which cannot freely diffuse in the system, usually because of its large size. The conditions lead to an unequal distribution of ions which are diffusible across a membrane separating the two volumes. In a repository, the non diffusible ions corresponds to the individual montmorillonite mineral layers, which are electrically charged and have a restricted mobility because of the relatively large size compared to the induced mean pore-size, see section 2.7.3, and the membrane corresponds to the rock interface where dissolved electrolyte ions may pass but not the montmorillonite layers Figure 2-12.

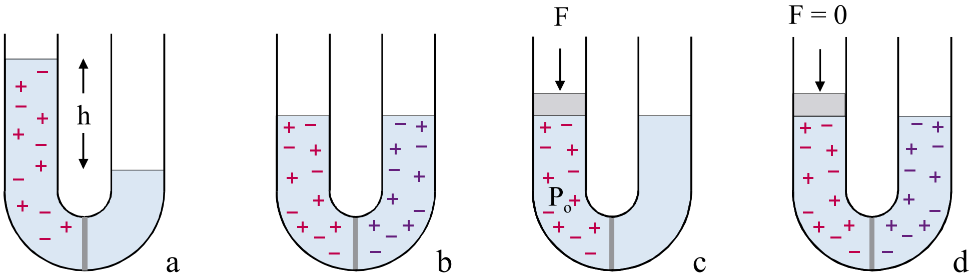

Figure 2-11. *The basic principles of osmosis as an analogue to bentonite swelling pressure.*

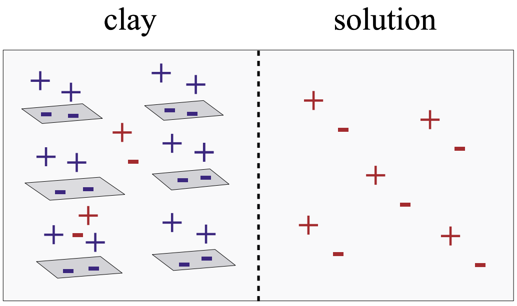

Figure 2-12. Principle drawing of the clay-water-ion system. Clay mineral flakes and charge compensating cations on the left side, and sodium and chloride ions on both sides of a semi-permeable membrane (buffer-rock interface), which is permeable to dissolved ions and water, but not to the individual montmorillonite layers.

The condition for equilibrium is that the chemical potentials are equal on both sides of the membrane for all components which can permeate. Since the macro-sized mineral layers cannot pass the membrane, the equilibrium will be established only for ions in the saturating solution and the montmorillonite charge compensating cations. Ideally, for sodium montmorillonite and a sodium chloride solution this will only be Na^+ and Cl^-. The conditions of electrical neutrality in the entire system are:

$$0 = z \cdot C_m + C_{Cl^-} - C_{Na^+}$$

where z is the valence of the macromolecule, C_m, C_{Na^+} and C_{Cl^-} is the concentration of the macromolecules, sodium ions and chloride ions, respectively. In terms of ion activity the equilibrium condition for the diffusible ions will be:

$$\{Na^+_c\}\{Cl^-_c\} = \{Na^+_e\}\{Cl^-_e\}$$

where the indices c and e refer to the clay-water system and to the surrounding electrolyte, respectively. The $\{Na^+_c\}$ factor is the sum of the concentration of original charge compensating ions $\{Na^+_{cc}\}$ and introduced electrolyte ions $\{Na^+_{ie}\}$. Local electrical neutrality has to be established thus $\{Na^+_e\} = \{Cl^-_e\}$ and $\{Na^+_{ie}\} = \{Cl^-_{ie}\}$ which give:

$$(\{Na^+_{cc}\}+\{Na^+_{ie}\})\ \{Na^+_{ie}\} = \{Na^+_e\}\ \{Na^+_e\}$$

Rearranging gives the introduced sodium ions:

$$\{Na^+_{ie}\} = \frac{-\left[\{Na^+_{cc}\} \pm \sqrt{\{Na^+_{cc}\}^2 + 4 \cdot \{Na^+_e\}^2}\right]}{2}$$

Consequently, it is possible to calculate the activity of sodium ions introduced into the clay-water system if the activity of the original cations $\{Na^+_{cc}\}$ in the clay and the activity of the ions in the external solution $\{Na^+_e\}$ are known. The resulting drop in swelling pressure can be approximately calculated since the activity of both the ground-water and the interlayer cations can be determined /Karnland et al, 2002/. Figure 2-13 shows measured and calculated results for the above conditions. The same principle may be applied to divalent ions, which leads to a more complicated expression, but both measured and calculated values for calcium show similar osmotic effects as for sodium.

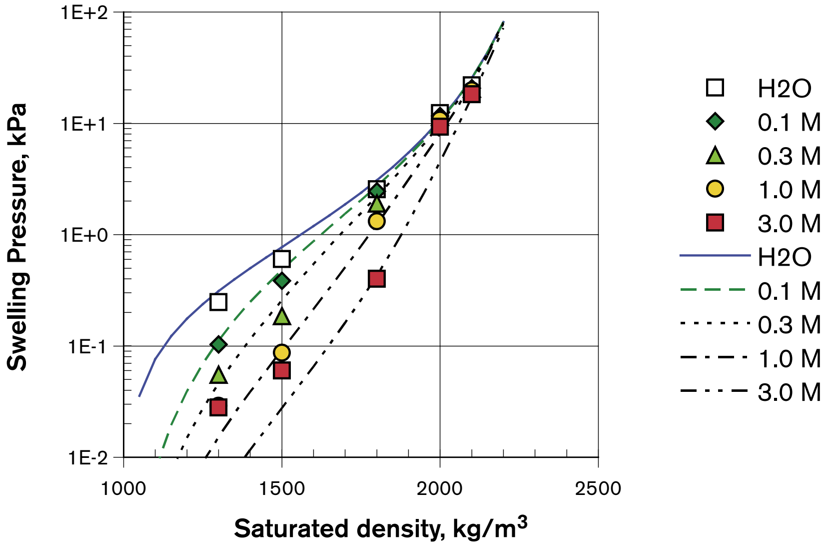

Figure 2-13. *Measured (squares) and calculated (lines) swelling pressure versus clay density for different concentrations in an NaCl solution in equilibrium with the Na-montmorillonite. Legends show external solution concentration in mole/L.*

The following table summarises how the process influences and is influenced by all buffer variables.

Variable/condition	Influencing (a)	Influenced (b)	Notes
Radiation intensity			
Temperature	no	yes	b) via gas law
Water content	yes	yes	a, b) primarily
Gas content	no	no	
Hydrovariables (pressure and flows)	yes	yes	by definition
Buffer geometry	yes	yes	a) via pore-water
Pore geometry	yes	yes	a, b) via pore-water
Stress state	yes	yes	a, b) via pore-water
Bentonite composition	no	yes	b) via pore-water
Montmorillonite composition	no	yes	b) via pore-water
Pore water composition	yes	yes	a, b) primarily
Structural and stray materials	No	No	

Boundary conditions

The process occurs at the interface between the bentonite and the outer solution. Generally, this is the rock/bentonite interface, but could also occur at the bentonite/canister interface in the case of a defect canister.

Model studies/experimental studies

The first preliminary study has been reported in SKB TR 1997. A comprehensive model and experimental study of MX-80 bentonite converted to a pure sodium state has been made by

SKB and is reported /Karnland et al, 2002/. An experimental study of the two reference bentonites MX-80 and Deponite-CAN is ongoing. Result for natural material, after conversion to pure sodium conditions, and after conversion to pure calcium conditions are presently available.

Time perspective

The process is relevant for all time perspectives in the lifetime of the repository. The effect of a change in ground-water salinity is expected to result in an almost instantaneous response in swelling pressure.

Natural analogues/observations in nature

Osmotic effects related to clay are discussed in several articles, but they are only peripherally relevant for repository conditions. Osmotic fluid pressure effects are recently discussed in /Neuzil, 2000/.

Handling in the safety assessment SR-Can

The osmotic swelling pressure effects are handled either by direct application of empirical data for relevant, scenario-specific conditions or by using the system evolution model, in e.g. calculations of buffer displacement as a result of swelling in saline groundwaters.

Uncertainties

Uncertainties in mechanistic understanding

The process is based on well established theory and generally well understood. Application to buffer conditions is however not clear in detail and quantification is not precise. The main remaining issue is the actual pore geometry, which is not determined in detail. At buffer conditions this may be relevant at very high ground-water conditions. For low clay densities, this issue is relevant for all ground-water concentrations.

Model simplification uncertainties for the above handling in SR-Can

The ion equilibrium model for predicting swelling pressure is based on the assumption that the system is relatively homogenous with respect to pore geometry. This assumption is supported by calculations of swelling pressure in electrolyte free montmorillonite by use of the Poisson-Boltzmann equation. If this assumption is not correct then the swelling pressure may decrease due to homogenization of the pore geometry with time.

Data uncertainties for the above handling in SR-Can

The input data to the model is the activity of ions in the ground-water, and the activity of the interlayer cations in montmorillonite. The model is relatively insensitive to the difference in activity and concentration for ground-water, and standard methods may be used for calculating this activity. The activity of the interlayer cations is not possible to determine by standard methods (Debey-Hückel, Davis, Pitzer etc). Uncertainties therefore still remain as to how this activity should be treated in a conservative way. A very conservative possibility is to use the same activity constants for interlayer ions as for the ground-water conditions. This will however not be applicable to the tunnel backfill material.

2.5.9 Montmorillonite transformation

Overview/general description

The advantageous physical properties of the buffer, e.g. swelling pressure and low hydraulic conductivity, are determined by the interaction between water and the montmorillonite mineral in the bentonite, see section 2.7.1. Minerals with the same principal structure but different layer charge occur in nature, see Figure 2-14. If the layer charge is near zero (pyrophyllite), there is virtually no interaction with water, which results in radically different properties than for montmorillonite. Minerals with higher layer charge and thereby more balancing cations may lead to greater interaction with water. However, the cation can be bound to the mineral surfaces if the layer charge is sufficiently high, and the interaction with water again ceases. The critical layer charge for cation fixation is dependent on the properties of the charge compensating cations. Layer charges above 1.2 unit charges per $O_{20}(OH)_4$, and potassium as counter-ion may result in no water interaction and fixation of the inter-lamellar distance to 1 nm. The end minerals, in which there is virtually no interaction with water, has 2 unit charges per $O_{20}(OH)_4$ for the potassium and sodium minerals (micas), and 4 units for calcium minerals (brittle micas).

At certain physico-chemical conditions transformation of montmorillonite may take place to some related minerals.

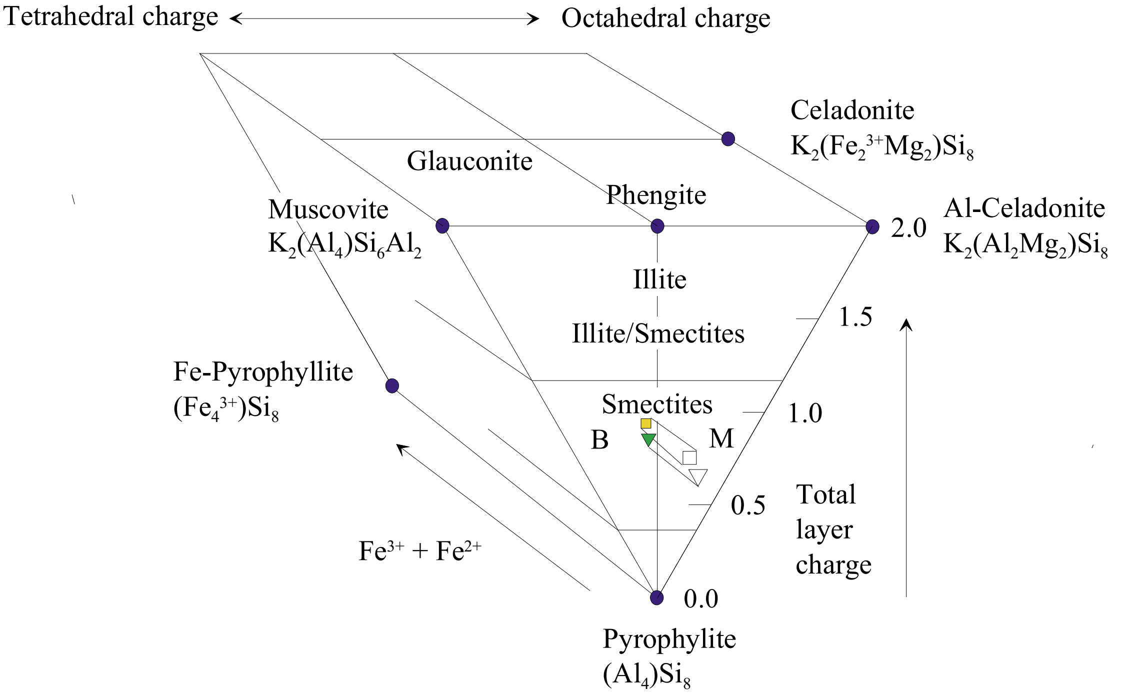

Figure 2-14. *Ideal end-member minerals in the pyrophyllite – mica series with potassium as charge compensating cation, and approximate compositional ranges for illite and smectite (modified from /Newman and Brown, 1987/). B denotes beidellite, M denotes montmorillonite. The compositional positions of the montmorillonite minerals in the reference bentonite materials are indicated by a triangle (MX-80) and by a square (Deponite-CAN). All formulas are related to the basic* $O_{20}(OH)_4$ *unit.*

Illitization

Transformation from montmorillonite to illite is well documented in several different geological formations, and has been reproduced under laboratory conditions. However, illite material is not well defined, and may be seen as a transition material from montmorillonite to mica minerals. All intermediate stages from swelling to non swelling material may be found (mixed layer smectite-illite) and several models have been suggested in order to describe the reaction. The conversion always involves a charge increase, mainly due to a decrease in silica content and an uptake of charge compensating potassium ions.

Illite material may be illustrated by the mean mineralogical composition of 24 natural illites /Weaver and Pollard, 1973/:

$(Si_{6.8}\,Al_{1.2})$	$(Al_{3.06}\,Fe^{3+}_{0.5}\,Mg_{0.56})$	$O_{20}\,(OH)_4$	$Ca_{0.1}\,Na_{0.06}\,K_{1.22}$
tetrahedral	octahedral		interlayer

which can be compared with the composition of the montmorillonite in the MX-80 SR-CAN reference bentonite:

$(Si_{7.86}\,Al_{0.14})$	$(Al_{3.11}\,Fe^{3+}_{0.37}\,Mg_{0.50})$	$O_{20}\,(OH)_4$	$Na_{0.47}\,Ca_{0.05}\,Mg_{0.02}\,K_{0.01}$
tetrahedral	octahedral		interlayer

The main differences being that the illites have approximately one unit charge higher tetrahedral charge, and potassium as the main charge compensating cations. It is generally accepted that a transformation may take place by release of silica from the montmorillonite tetrahedral sheet, and that no complete dissolution of montmorillonite is necessary. Simplified, the total illitization reaction may be expressed:

$$Ca^{2+}/Na^{+}\text{-montmorillonite} + K^{+} + (Al^{3+}) \rightarrow \text{Illite} + \text{Silica} + Ca^{2+}/Na^{+}$$

In massive natural sediments the physico-chemical conditions are similar over huge volumes, and concentration gradients are generally insignificant. The total reaction therefore involves a series of sub-reactions; release of silica from montmorillonite, release of potassium from e.g. feldspars, potassium fixation and precipitation of new silica minerals, and the total reaction rate is governed by the slowest of the sub-reactions.

According to the equilibrium conditions for 25°C shown in Figure 2-15, the montmorillonite will be thermodynamically stable for silica activities over 1 mM, and silica has to precipitate or be transported away in order to let an illitization reaction take place. Amorphous silica, with an equilibrium activity of around 2 mM, may consequently prevent the dissolution of silica from montmorillonite, but quartz may not. Quartz precipitation may therefore be a kinetically controlling mechanism in illitization of natural montmorillonite sediments.

Kinetic models have been developed for calculation of degree of transformation in smectite-to-illite transformation. Relationships and constants have been established by means of laboratory batch experiments and comparisons with natural systems /e.g. Eberl and Hower, 1976; Pytte, 1982; Huang et al, 1993/. Most models have the form of an Arrhenius expression and the Huang et al model may be shown as an example:

The overall kinetics of the smectite-to-illite reaction can be described by the equation:

$$-dS/dt = A \cdot [K^{+}] \cdot S^{2} \cdot \exp(-E_a/RT),$$

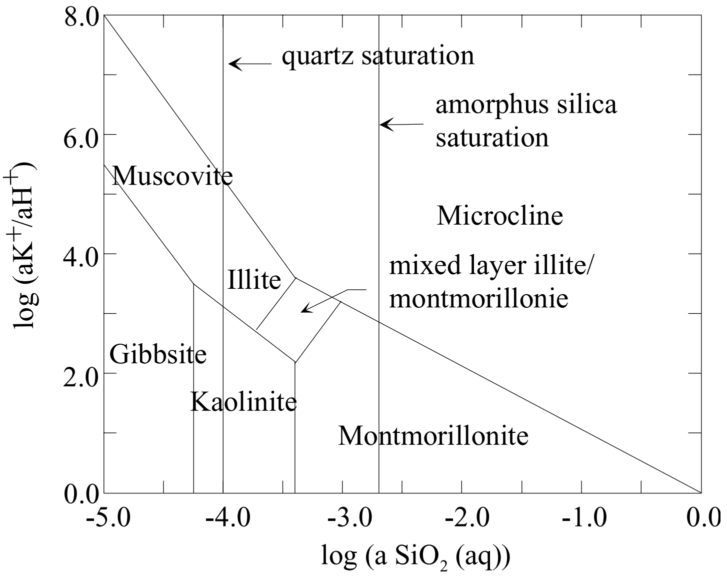

Figure 2-15. *Silica mineral equilibria with the partial component potassium at 25°C. (Simplified from /Aagard and Helgeson, 1983/).*

where S is the smectite fraction in the illite/smectite material, A is frequency factor, E_a is activation energy and R is the universal gas constant, and T is temperature. After integration of Equation 2-1, the smectite content at a certain time can be calculated if the temperature and potassium concentration in the pore water are known. The potassium concentrations in the Äspö groundwater are measured to be in the range of a few ppm up to 80 ppm /Nilsson, 1995/. According to the model, practically no clay conversion is possible in a KBS-3-type repository at these conditions as shown in Figure 2-16.

Rectoritization

Rectorite may be seen as a sodium equivalent to illite. It is much more rare in natural systems compared to illite. The mean layer charge is higher than in illite, normally close to 2 units charges per $O_{20}(OH)_4$. Transformation of montmorillonite into rectorite is theoretically possible if potassium is not available, and release of silica from the montmorillonite continues until the required tetrahedral charge is reached.

Chloritization

Chlorites have the same basic structure as montmorillonite, but the layer charge is normally close to 2 units charges per $O_{20}(OH)_4$. The charge is balanced by positively charged octahedrally coordinated hydroxide sheets. The central ion in the interlayer hydroxide sheet may be any di- or tri-valent metal ion, normally Mg, Al or Fe. There are consequently two octahedral sheets in a unit structure, and no exchangeable interlayer cations. Chlorites are less commonly found as alteration products from montmorillonite in natural sediments compared to illites, and the chlorites are generally formed at higher temperatures. Transformation of montmorillonite into chlorites is theoretically possible if potassium is not available, but di- or trivalent cations are, and if release of silica from the montmorillonite continues until the required tetrahedral charge is reached.

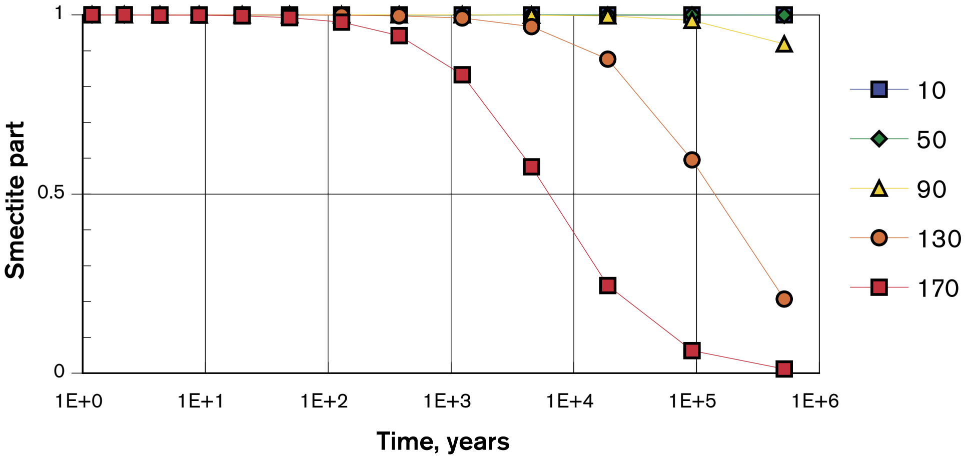

Figure 2-16. *Remaining smectite part for different temperatures in a hydrothermal system with* $[K^+] = 0.002$ *mole/litre (80 ppm) according to the Huang et al kinetic model and laboratory determined constants* ($E_a = 27.4$ *kcal/mole and* $A = 8.5E4$) */Karnland, 1995/. Legend shows temperature in °C.*

Reduction of octahedral iron

The octahedral layer in montmorillonite frequently contains a significant amount of iron. Normally, the iron is in an oxidized condition in commercial material due to exposure to air during mining and handling. Reduction to Fe(II) would lead to an increase of the total layer charge, and a potential fixation of interlayer cations if the charge change is sufficient. The effect of such octahedral layer change is expected to have less effect compared to a similar charge change in the tetrahedral layer /Spositio, 1999/. The SR-Can reference bentonites contain 0.37 Fe (MX-80) and 0.45 Fe (Deponit CA-N) per $O_{20}(OH)_4$ unit, and a total reduction of the iron would give a layer charge close to the illitization charge limit.

Dissolution of silica due to high pH

The solubility of silica minerals increases strongly with increasing pH, see Figure 2-17. Increased dissolution of tetrahedral silica in montmorillonite will take place parallel to dissolution of other silica minerals at high pH. If a pH gradient prevails in the system the resulting activity gradient will lead to significant transport of dissolved silica, and a charge change in the tertahedral layer in the montmorillonite. At stagnant conditions this will result in formation of new minerals stable under the prevailing high pH conditions. Such reactions are not covered by the kinetic expressions for illitization.

Effects of transformation

Change in the layer charge of the montmorillonite leads to change in the interaction with water and thereby to effects on the swelling pressure. On illitization, the interaction virtually ceases and the water previously bound between the individual layers is released. The pore geometry is thereby changed towards fewer and bigger pores. Swelling between remaining montmorillonite layers will partially balance the negative effects. The quantity of remaining montmorillonite will thereby determine the system's swelling pressure and hydraulic conductivity. The sensitivity to the ion content of the pore water increases with the degree of transformation. A total transformation of the montmorillonite into illite or chlorite would lead to complete loss of buffer functions.

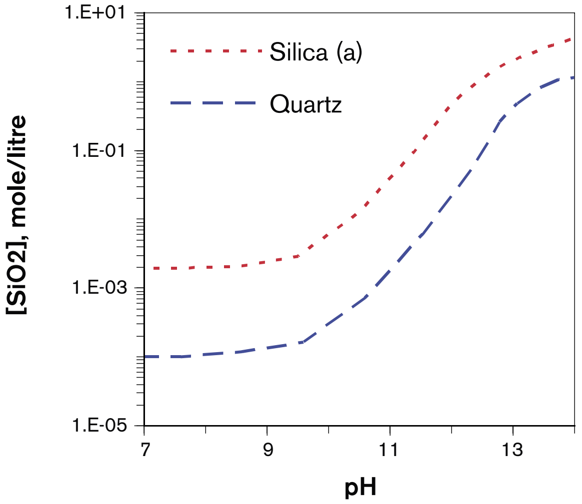

Figure 2-17. *Silica equilibrium activity conditions for amorphous silica and quartz versus pH as calculated by PHREEQC / Lawrence Livermore database.*

Released silicon is expected to lead to a deterioration of the buffer's rheological properties.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable/condition	Influencing (a)	Influenced (b)	Notes
Radiation intensity	no	no	b) possibly
Temperature	yes	yes	a) change in lambda
Water content	yes	no	via buffer geometry
Gas content			
Hydrovariables (pressure and flows)	yes	yes	
Buffer geometry	yes	yes	
Pore geometry	yes	yes	via pore-water
Stress state	yes	yes	via pore-water
Bentonite composition	yes	yes	b) via pore-water
Montmorillonite composition	yes	yes	by definition
Pore water composition	yes	yes	a) primarily
Structural and stray materials			

Boundary conditions

There are no particular boundary conditions to discuss for this process. The relevant boundary conditions in order to treat the process quantitatively are those of the transport processes that control the exchange of solutes between the buffer pore water and the water in its adjacent components, i.e. the boundary conditions of the processes diffusion and advection.

Model studies/experimental studies

The illitization process is of great importance from several viewpoints in connection with prospecting and extraction of oil deposits. The great commercial interest, in combination with the mineralogically interesting process, has led to very extensive research since the 1960s. A large number of publications therefore exist as a basis for the assumption that the illitization process is not expected to be important in a deep repository. The argumentation can be summarized in three points:

1. The bentonite material is close to mineralogical equilibrium to start with.

   In its original environment, the bentonite normally comprises a system that is close to mineralogical equilibrium, i.e. only insignificant mineralogical changes take place. In a deep repository, the exchange between buffer and surrounding environment is strongly limited, and the mass of the buffer is relatively great. The bentonite in the buffer will therefore create its own chemical environment to start with /see e.g. Fritz et al, 1984/. The system can thus be expected to be close to mineralogical equilibrium during most of the repository period.

   Two early changes in the chemical environment are obvious, however:
   - Increase in the ion content of the pore water as a result of inward transport of solutes during water saturation. For ordinary groundwater, the increase is small in relation to the naturally high cation concentration that exists in the montmorillonite in the buffer.
   - Temperature increase as a result of fuel decay. The period with significantly elevated temperature is, however, relatively short (<5,000 years) /Thunvik and Braester, 1991/.

2. Temperature-induced transformation is limited by a shortage of K^+.

   In order for a transformation to illite to take place, the charge increase must be balanced by potassium ions. Since the natural fraction of potassium in the buffer may be kept low, potassium must be transported in from the surroundings. The potassium requirement to completely convert the montmorillonite in the buffer is about five weight-percent, which is equivalent to nearly a tonne of potassium for a deposition hole. Calculations and modellings show that the inward transport is expected to take place very slowly and that this in itself constitutes an effective obstacle to a substantial transformation /Hökmark, 1995/.

3. The transformation rate at the repository's maximum temperature is very low.

   Kinetic models have been developed for calculation of degree of transformation in smectite-to-illite transformation. Relationships and constants have been established by means of laboratory experiments and comparisons with natural systems /e.g. Eberl and Hower, 1976; Pytte, 1982; Huang et al, 1993/.

Experimental verifications: Since the processes are very slow, laboratory experiments must be conducted under accelerated conditions, i.e. at high temperatures and potassium concentrations, in order to obtain measurable results /Karnland et al, 1995/. Comparisons with natural transformations provide an opportunity to verify laboratory results /Elliott and Matisoff, 1996/.

The mineralogical stability is studied in the Lot tests at Äspö HRL. The test conditions represent repository conditions and adverse conditions in order to accelerate possible degradation processes. The adverse conditions involve:

- Higher temperature.
- Higher temperature gradient.

- Higher content of accessory minerals (calcite, gypsum, K-feldspar).
- Introduction of new substances (Portland cement).

The expected effect of these accelerating conditions (buffer adverse conditions) may be summarised in the following discussion.

The initial reaction rate of a reaction

> $aA + bB \rightarrow$ products
>
> can be described by the standard rate law
>
> Rate = $k\,[A]^n\,[B]^m$
>
> where n and m are constants, and k is a rate constant which can be expressed by the Arrhenius relationship
>
> $k = zp \cdot \exp(-E_a/RT)$,
>
> where z is the collision frequency factor, p is the steric factor, and E_a is activation energy and R is the gas constant.

This reaction rate theory consequently implies:

- Reaction rate increases with increasing concentration of a reactant.
- Reaction rate increases with increasing temperature.
- Temperature increase effect is reduced with increasing temperature.
- Temperature effect increases with increasing activation energy.

A slow reaction rate is normally due to high activation energy, which means that the effect of temperature is more pronounced for slow reactions. E.g. in the case of illitisation a rather high activation energy is measured by Huang et al (114700 J/mole), and a temperature increase of 10 K results in a close to tripled reaction rate. The increase from maximum temperature in the KBS-3 buffer to the maximum temperature in the adverse conditions parcels (130°C) leads to an increased illitisation reaction rate of around 40 times. Hundred years after deposition the most conservative assumption concerning water saturation gives a calculated maximum temperature increase of around 60°C in a KBS-3 repository, and the corresponding temperature after 1000 years is 40°C. A comparison between the 130°C in the adverse condition parcel and a constant bentonite temperature of 70°C (60°C increase plus background) gives a reaction rate factor of almost 400 times. In a 5-year test, only the effect of increased temperature in the A parcels will accordingly simulate the major period of elevated temperature in a KBS-3 repository, with respect to kinetically governed processes.

Consequently, the Lot tests can be expected to show the effects of processes governed by the kinetics of the involved reactions. The introduction of additional substances and increased temperature gradients is made in order to accelerate also processes, which are governed by transport of substances. The effect of the temperature gradient is, however, not possible to describe in an, as simple and general way, as the effect of increased temperature and concentrations of reactants.

Natural analogues/observations in nature

Besides in SKB-related investigations, the transformation of smectite to illite is well-documented for a large number of geological formations. Natural sediments have been studied by, among others, /Burst, 1959; Perry and Hower, 1970; Hower et al, 1976; Colten-Bradley, 1987; Lynch, 1997/. The studies show that increased sediment depth (increasing temperature) leads to increased illite content, and that low availability of potassium can be linked to reduced transformation. Silica activity as kinetically controlling factor has been studied in sediments /Abercrombie et al, 1994/. Special formations where temperature effects have been obtained from e.g. volcanic activity have revealed a similar transformation /e.g. Pytte, 1982; Lynch, 1985; Brusewitz, 1986/. Cementation effects as a consequence of transformation have been studied specifically by, among others, /Hower et al, 1976; Boles and Franks, 1979/.

Time perspective

Precipitation and montmorillonite layer charge changes may take place during the initial temperature gradient phase.

Significant changes in the montmorillonite content of the buffer are expected to occur on a million-year timescale according to illitization models.

Handling in the safety assessment SR-Can

Temperature: The maximum temperature will tentatively be modelled by the kinetic models for illitization for different realistic concentrations of potassium. The temperature gradient during the initial phase is more problematic. The conditions have to be modelled by an equilibrium/transport code with respect to silica for different interaction with groundwater. The maximum possible transport of silica may be used to quantify the maximum charge effect on montmorillonite, and thereby on all transformation processes except dissolution /precipitation reactions, which is covered by the kinetic models. Based on the presently available models for illitization, a temperature of 100°C would give negligible illitization.

pH: The effect of high pH will be modelled by a reactive/transport code. The maximum release of silica from montmorillonite may be used to quantify the transformation. Based on presently available data, mainly from the ECOCLAY project, the alkalinity of solutions in contact with the buffer should not exceed pH 11.

Uncertainties

Uncertainties in mechanistic understanding

There is a generally accepted explanation to the diagenesis in which smectite is converted into illite accompanied by a release of silica, water and cations. The main reaction may be expressed as:

$\text{Smectite} + K^{+} + (Al^{3+}) \rightarrow \text{Illite} + Si^{4+} + Ca^{2+}/Na^{+}$

This smectite-to-illite transition has been reproduced under laboratory conditions and has been identified in the following other geological situations:

- smectite-bearing shales intruded by dikes, which yield a transition series along a line perpendicular to the surface of the dike /Lynch and Reynolds, 1985/,
- geothermal areas, which are developed farther than what have been found in burial diagenesis /Jennings and Thompson, 1986/,

- hydrothermal zones around ore bodies, which shows similar but more complex series /Horton, 1985/.

There is, however, no general consensus concerning the details in the smectite-to-illite transformation mechanisms and a number of explanations of the character and mechanisms of the smectite-to-illite conversion have been proposed. The following descriptions of four models may serve as an illustration of different views /Moore and Reynolds, 1989/:

- MacEvans crystallite model,
- fundamental particle model,
- segregated model,
- two-solid-solution model.

In the MacEvans model the layers of illite and smectite are intimately interlayered, probably epitaxially connected, stacked either randomly or regularly, into a fixed sequence. The transition from smectite into illite is made without disrupting the stacks of layers. A layer with smectite chemistry and structure is changed in place into an illite.

The second model created by Nadeau and his coworkers /Nadeau et al, 1984/, have three types of fundamental particles: an illite or K-unit, a smectite unit and a chlorite unit. The silicon skeleton is thereby irrelevant. If the particle is 10 Å thick it is a smectite particle. Illite particles are at least 20 Å thick and are composed by two smectite particles held together by interlayer K. Finally, the chlorite particles are about 24 Å thick and are composed by two smectite particles joined by a (Mg, Fe) hydroxide sheet in the interlayer space. The external surfaces of these three types of fundamental particles will adsorb water or other polar molecules. This model suggests that the particles are not grown together epitaxially. The sequence of layers is not fixed in relatively large crystallites but simply stacked together into a group of particles which are enough parallel to reflect X-rays.

The segregated model is based on images made with a high resolution transmission electron microscope (HRTEM) by Peacor and his coworkers studying illite/smectite samples. They interpreted straight, defect-free layers as illite and wavy layers as smectite. They proposed two mechanisms for the formation of illite in the smectite-to-illite transition. In the first case, illite and smectite exists distinctly separated from each other but in the same relatively large packlets of layers. In this type of illite formation, which they think, is characteristic of the US Gulf Cost sediments, packets of illite are seen within layers of smectite, indicating that the illite has inherited structures from the smectite. In the second case, characterized by samples from the Salton Sea geothermal field, illite crystals are physically quite distant from smectite on a microscopic scale and have a pseudohexagonal shape. /Yau et al, 1987/ inferred that these illite crystals had been precipitated from solution.

The last model described is called the two-solid-solution model and is based on the work made by /Inoue et al, 1987/. They identified three phases in the hydrothermally altered system they studied: two are solid solutions and the other shows little variation in chemical composition. Of the two solid solutions, one varies from 100 to 50% expandable layers with random stacking. The other varies from about 50% expandable layers to pure illite, with regularly to partially ordered stacking. The first solid solution is characterized as smectite undergoing K-fixation, and the second as maturing illite. The third phase present has less than 5% expandable layers and a distinct morphology. The morphology of the three phases were studied by use of TEM photomicrographs and showed three different distinct shapes. The conclusions are that the illite laths are increasing in size by the addition of material from solution and that the material is coming from smectite flakes that are decreasing in number because they are dissolving. This inference is supported by chemical analyses that show coexisting flakes and laths to have the same interlayer K content, and the fact that flakes are rare in samples showing more than 50% illite. They proposed two different reactions for these two series. K-fixation takes place as a remodelling or transformation in the flakes, whereas the laths grow because the flakes are dissolving and being re-precipitated as laths or as additions to laths.

Kinetic aspects

Beside the large number of studies concerning smectite-to-illite reaction in nature, numerous experiments have been carried out hydrothermally in laboratory. Most of these experiments have focused on certain issues such as the effect of interlayer cations /Eberl, 1978/, tetrahedral substitution /Huang and Otten, 1987/, octahedral substitution /Güven and Huang, 1991/, solution chemistry /Roberson and Lahann, 1981/, kinetics of layer charge development /Howard and Roy, 1985/, control of ordering of mixed-layers /Huang, 1989/, and the effect of flow rate in flow through experiments /Kacandes et al, 1991/.

Several of these models suggests that the conversion rate can be calculated by use of an expression based on the Arrhenius equation. The models are though somewhat different in character. /Eberl and Hower, 1976/ proposed a first order kinetic equation based on experiments with synthetic beidellites. This model is however not applicable to field observations because the illitisation rate of this synthetic smectite was found to be significantly different than that of natural smectite /Huang and Otten, 1987/. /Bethke and Altaner, 1986/ derived a theorethical rate law by assuming the layer-by-layer substitution mechanism of smectite-to-illite transformation. /Pytte and Reynolds, 1989/ derived a six order kinetic equation based on the field observations of progressive illitization of smectite in a contact methamorphic zone. This model has been widely used for modeling natural illitization processes. /Velde and Vasseur, 1992/ proposed an empirical kinetic model based on empirical field data from three sedimentary basins. /Wei et al, 1993/ proposed a model which takes into account an activation energy distribution in the smectite-to-illite reaction depending on variations in the original smectite mineral. /Huang et al, 1993/ used natural smectite in order to quantify the kinetics of the smectie-to-illite reaction experimentally. The conversion rate could be described by a simple rate law that is second-order with respect to the fraction of smectite and first-order with respect to K^+ concentration. Since the Huang et al model is based only on laboratory tests and successfully tested on geological smectite-to-illite reactions it may be used in order to model possible reactions in a repository.

For obvious reasons, the kinetic model constants are determined in laboratory experiments performed with relatively short duration time and at high temperatures and high potassium concentrations compared to the natural conditions it is aimed to describe. In order to use the proposed model for repository conditions, the following issues are of importance:

- how well does the relationship describe the test reaction,
- how well determined are the constants, and
- is the model sufficiently generally applicable in order to be used for repository conditions?

An argument for the Huang model, compared to others, is that it is consistent with the principles of chemical kinetics /Lasaga, 1981/, which suggests that the rate of conversion is, in general, proportional to the activity products of the reactants. In other words, the model predicts two smectite ”molecules” to react with one K^+ ion to form one ”molecule” of illite. The first order with respect to potassium concentration is also consistent with the experimental data.

Model simplification uncertainties for the above handling in SR-Can

The existing models for illitization deal with the total reaction from montmorillonite to illite. Most experiments and natural analogues represent stagnant conditions, and thereby kinetic controlling mechanisms which are not present under the initial heating phase with temperature gradients. In the first transient heating phase of the repository, the temperature gradient will lead to a corresponding silica concentration gradient and diffusive transport of silica from the canisters towards the rock. The release of silica from the montmorillonite (i.a. illitisation reaction) may consequently be faster in an open system where transport of silica may be possible.

The subsequent phases may also depart from the illitization model conditions in that sense that the exchange with ground-water may be governing the reaction, i.e. silica release. This is then governed by the silica concentration in the ground-water.

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

Thermodynamic and kinetic data for modelling silica dissolution/precipitation are not well known especially for high pH and high temperatures. In transport calculations, the diffusion constants in rock, bentonite and bentonite constitute an uncertainty.

2.5.10 Colloid release

Overview/general description

The water uptake into the bentonite buffer is counteracted by the walls in the deposition hole, and a swelling pressure is developed in the bentonite (see section 2.6.1). Fractures intersecting the deposition hole imply that no swelling restrictions are present, and that swelling continues into the fractures until a thermodynamic equilibrium is reached without the development of swelling pressure. This free swelling may lead to separation of individual montmorillonite layers, or small groups of mineral layers (dispersion).

The maximum free swelling is strongly dependant on the salinity of the ground-water. The interlayer distance may be discussed in terms of the Debey-Hückel screening length theory, which predicts a spacing change inversely proportional to the square root of solution concentration, which has been experimentally confirmed.

At low concentration the distance between the individual montmorillonite layers may be large enough to let the clay particles be dominated by thermal motion i.e. they have colloidal properties. The individual colloidal particles may then be transported away from the deposition hole in fractures by diffusion or by ground-water flow in the fracture.

In general, the stability of such a colloidal system can ideally be evaluated from DLVO theory in which all attraction and repulsive forces between individual montmorillonite layers are taken into account. Coagulation of a dispersed system may be achieved by adding an electrolyte, and the higher the valence, the more efficient is the coagulation. A critical coagulation concentration (CCC) may be determined for different solutions, and this CCC-value may be used as a conservative value for preventing swelling to an extent where montmorillonite will appear as a colloid.

The critical coagulation concentration (CCC) may be determined for different solutions, and used as a conservative value for spontaneous colloid formation. Ground-water concentration has to be at least 0.1 M with respect to sodium (monovalent) ions or 0.001 M calcium ions (divalent ions) in order to neglect colloid formation. If this is not achieved mass transport modelling, including diffusive and flow transport, has to be made.

The following table summarises how the process influences and is influenced by all buffer variables. Note though that the process occurs at the boundary between buffer and rock, within rock fractures making the boundary conditions described below of prime importance for the understanding and quantification of the process.

Variable/condition	Influencing (a)	Influenced (b)	Notes
Radiation intensity	No	no	
Temperature	Yes	yes	
Water content	Yes	yes	via pore-water
Gas content	No	no	
Hydrovariables (pressure and flows)	Yes	yes	
Buffer geometry	Yes	yes	
Pore geometry	Yes	yes	a, b) via pore-water
Stress state	Yes	yes	a, b) via pore-water
Bentonite composition	Yes	yes	a, b) via pore-water
Montmorillonite composition	No	yes	b) via pore-water
Pore water composition	Yes	yes	a, b) primarily
Structural and stray materials	No	No	

Boundary conditions

The boundary conditions are the cation concentration and cation valency of the groundwater in direct contact with the outermost part of the bentonite buffer.

Water flow in the fractures will not affect the stability of true colloids. However, the interlayer distance between the montmorillonite layers is continuously increasing with decreasing electrolyte concentration and layers at large separation are more sensitive to erosion than layers at close separation. Water-flow may consequently tear loose montmorillonite layers although the distances are shorter than what is needed for true colloids. The extent of colloid formation by flow is consequently dependent on the flow velocity, and the colloids are not stable without flow. The treatment of erosion is treated in section 2.5.4.

Model studies/experimental studies

The principles for swelling of montmorillonite in electrolyte solutions have been reported in a large number of publications. The basic theoretical principles are described by the DLVO theory /Verwey et al, 1948; Derjaguin and Landau, 1941/. Interlayer distances were experimentally determined in by /Norrish and Quirk, 1954/, and several subsequent studies have in principle confirmed these results. Several laboratory studies have been made in order to determine the critical coagulation concentration for montmorillonite in sodium and calcium chloride solutions /van Olphen, 1963; Le Bell, 1978; Wold, 2003/.

Natural analogues/observations in nature

In principle, a large number of analyses of Swedish ground-water may serve as natural analogs since they show low contents of clay colloids despite the fact that swelling clay is often found in fractures. Specific studies have been made at the Äspö HRL /Laaksoharju, 2003/ and at Grimsel HRL /Degueldre et al, 1996/.

Time perspective

Colloid formation is only relevant at low ion concentrations and likely only relevant inonjunction with a glaciation. The effects will be significant only in the long term perspective.

Handling in the safety assessment SR-Can

The process is neglected if the sum total concentration of divalent cations exceeds 1 mM. If this condition is not fulfilled, loss due to erosion needs to be quantified. A first version of a model for such a quantification is presented in Appendix A.

Uncertainties

Uncertainties in mechanistic understanding

The knowledge concerning colloid formation and colloid stability is good concerning the effects of mono- and divalent ions. The effect of trivalent ions is theoretically significantly larger than for divalent ion and could possibly serve as a less conservative criterion. The effects are not well established in laboratory experiments for the actual conditions. The possible transport of colloids by diffusive and flow transport is less well known. The effects of mechanical erosion of loose gels close to colloid conditions have not been examined.

Model simplification uncertainties for the above handling in SR-Can

The suggested treatment in SR-Can is based on a simplified, scoping model (Appendix A). It should be noted that a number of simplifying assumptions have been made.

The following assumptions must be revisited and checked for reasonableness

1. The change of composition in the diffuse layer is not accounted for.
2. Steady state assumption for clay intrusion into the fracture.
3. Steady state assumption of the chemical transport.
4. Applicability of Q_{eq} model.
5. Assumption that the Ca transport in the bulk of the clay in the deposition hole can be neglected.
6. Assumption of no mechanical erosion of gel fringe.
7. Assumption of Ca mineral dissolution.
8. Assumption of steady state for chemistry- e.g. no pH changes that influence the CCC.

The above items can all conceivably influence (increase) the dispersion rate. There may be more that will be found in a more detailed investigation

The following could act to decrease the dispersion rate

1. The diffusion resistance in the clay in the fracture is neglected.
2. The change of composition in the diffuse layer is not accounted for.
3. Physical hindering of clay particle dispersion in the fracture.
4. Clogging of water pathways by the clay being pressed out into the fracture.

5. Sedimentation of clay particles in fracture.
6. Development of a montmorillonite depleted region near the fracture entrance leaving behind sparsely soluble minerals that “clog” the fracture entrance.

There may be more effects that will be found in a more detailed investigation

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

The evolution of ground-water composition, and ground-water flow conditions are the important uncertainties. These will be evaluated in integrated assessment modellings, see further the Interim report of the safety assessment SR-Can.

2.5.11 Radiation-induced transformations

Overview/general description

Montmorillonite in the buffer can be broken down by γ radiation. The result is a decrease in the montmorillonite concentration.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	Yes	
Temperature	No	No	
Water content	No	No	
Gas content	No	No	
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	No	No	
Pore geometry	No	No	
Stress state	No	No	
Montmorillonite composition	No	?	Not detected
Pore water composition	No	No	
Bentonite composition	No	?	Not detected
Structural and stray materials	No	No	

Boundary conditions

A quantitative treatment of the process requires knowledge of the radiation field in the buffer, which is obtained from the process Radiation attenuation, section 2.3.1.

Model studies/experimental studies

Experimental studies have shown that the accumulated radiation doses to which the bentonite will be exposed in a deep repository do not cause any measurable changes in the montmorillonite concentration. MX-80 bentonite saturated with weakly brackish water to a density of 2,050 kg/m^3 was irradiated for one year with a total radiation dose of $3 \cdot 10^7$ Gy, which is orders of magnitude more than what will be the case in the repository. The specimen was confined in a cylinder whose one closed end consisted of iron and irradiated with a ^{60}Co source, while the other end consisted of a porous steel filter through which a water pressure of 1.5 MPa acted /Pusch et al, 1993/. The irradiated end, which had an adsorbed dose rate of 3,972 Gy/h, was kept at 130°C, and the opposite end, which had a dose rate of 456 Gy/h, at 90°C.

Mineral assays (XRD, IR, CEC) of this specimen and of a parallel specimen not exposed to radiation but to the same hydrothermal environment as the irradiated specimen, showed no significant change in the montmorillonite quantity in either of the specimens.

Natural analogues/observations in nature

Not applicable.

Time perspective

See section 2.3.1.

Handling in the safety assessment SR-Can

Based on the experimental studies mentioned above, the process is neglected.

Uncertainties

There are no uncertainties of relevance to the safety assessment.

2.5.12 Radiolysis of pore water

Overview/general description

Gamma radiation from the fuel that penetrates through the canister can decompose pore water by radiolysis, forming OH radicals, H_2, O_2 and several other components. The oxygen is consumed rapidly by oxidation processes which affect the redox potential, while the hydrogen is transported away. The canister's wall thickness is, however, sufficient so that the effect of γ-radiolysis on the outside is negligible /Werme, 1998/.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	Yes	
Temperature	No	No	
Water content	No	Yes	
Gas content	No	(yes)	
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	No	(yes)	
Pore geometry	No	No	
Stress state	No	No	
Montmorillonite composition	No	No	
Pore water composition	Yes	Yes	
Bentonite composition	No	No	
Structural and stray materials	No	No	

Boundary conditions

A quantitative treatment of the process requires knowledge of the radiation field in the buffer, which is obtained from the process Radiation attenuation.

Model studies/experimental studies

Corrosion of copper in water in the presence of g-radiation has been studied I a few experiments. These a summarized in /King et al, 2001/. The results show no effects on the corrosion rate even with dose rates much higher than those expected in the repository.

Natural analogues/observations in nature

Not applicable.

Time perspective

The γ radiation is significant during approximately 1,000 years.

Handling in the safety assessment SR-Can

Radiolysis of the porewater could potentially increase the corrosion rate of the copper canister. The experiments mentioned above clearly show that this is not the case. The process is therefore neglected in SR-can

Uncertainties

There are no uncertainties of relevance to the safety assessment.

2.5.13 Microbial processes

Overview/general description

Microbial processes can under certain conditions result in the formation of gas and sulphide /Pedersen, 2002/. Gas formation can give rise to disrupting mechanical effects on the buffer, and sulphide from sulphate reducing bacteria (SRB) can corrode the copper canister. Sulphide formation must take place in the buffer, near the canister, and be of considerable scope for corrosion to be possible, mainly due to the fact that the solubility of sulphide, and thereby its diffusive transport capacity, is very low. In order for the above-described processes to take place, the bacteria must be active and have access to water, nutrients and space /Pedersen, 2002/.

Sulphate reducing bacteria are very common in deep Fennoscandian groundwater. They are reported to occur abundantly down to depths of at least 600 to 700 m /Haveman and Pedersen, 2002; Pedersen, 2000b/. Microbes are generally very robust and can withstand harsh conditions. The potential for sulphide production is significant at repository depth and possible effects on the repository function must be considered in safety assessment.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No effect	Microbes are generally radiation resistant, but it varies from species to species.	Increasing dose will decrease the number of surviving microbes.
Temperature	No effect	All microbes have a temperature range within which they are active. Increasing the temperature will increase microbial activity.	Temperatures above the max are lethal. Highest know temperature for microbial life is about 110–120°C
Water content	No effect	Decreasing water availability reduces the diversity. Many microbes can compensate a low water content using metabolic, energy consuming processes.	Water is needed for active life. A combination of low energy availability and a low water content is detrimental to many microbes
Gas content	Active microbes can produce gas if organic carbon is available for metabolism.	Hydrogen and methane can be consumed by microbes; best effect is obtained if oxygen is present.	Microbes both produce and consume gases
Hydrovariables (pressure and flows)	No effect	Most microbes tolerate large variations in hydrostatic pressure and flow.	
Buffer geometry	No effect	No effect	
Pore geometry	No effect	Pores with sizes smaller than about 200 nm can not be penetrated or inhabited by microbes	Microbes vary in size from about 200 nm up to 600 μm.
Stress state	No effect	Microbes tolerate limited mechanical pressure. Swelling clay will disrupt microbes when the pore size becomes smaller than the size of the microbe.	
Montmorillonite composition	No effect	No known effect.	
Pore water composition	Limited effect if the microbes are metabolically active. Gas and sulphide production can theoretically occur.	There is a positive correlation between microbial activity and the concentration of organic carbon.	This will only be important if the degree of compaction is low enough to allow microbial activity.
Bentonite composition	No effect	There is a positive correlation between microbial activity and the concentration of organic carbon.	
Structural and stray materials	No effect	No known effect.	

Boundary conditions

There are no particular boundary conditions to discuss for this process. The relevant boundary conditions in order to treat the process quantitatively are those of the transport processes that control the exchange of solutes (microbe nutrients) between the buffer pore water and the water in its adjacent components, i.e. the boundary conditions of the processes diffusion and advection.

Model studies/experimental studies

A Large scale Canadian buffer experiment

A full-scale experiment with buffer material consisting of 50/50% bentonite/sand was performed at Atomic Energy of Canada Limited´s (AECL) underground laboratory in Canada. The results showed that microbes, with a few exceptions, could only be cultured from buffer samples with a water content of 15% or more, which is approximately equivalent to a 100% bentonite density of 2000 kg/m^3 /Stroes-Gascoyne et al, 1997/. Elevated temperatures had no effect on the microbes. These results were interpreted as an effect of limited availability of water. The result of the Buffer Mass Container (BMC) experiment invoked questions about the survival of microbes, and especially SRB, in buffer materials in 100% bentonite and led to detailed laboratory experiments.

Laboratory experiments exploring the survival of bacteria in buffer

The Canadian field experiment was followed up by laboratory experiments. Two species of sulfate reducing bacteria were mixed with MX-80 bentonite at varying densities, from 1500 kg/m^3 to 2000 kg/m^3 /Motamedi et al, 1996/. The species were Desulfovibrio aespoeensis and Desulfomicrobium baculatum, both isolated from deep groundwater at the Äspö HRL. None of the species survived 60 days at densities above 1800 kg/m^3. Desulfomicrobium baculatum survived the better of the two, remaining culturable for 60 days at 1500 kg/m^3. Although microbial activity rapidly diminished, it was argued that this was due to the laboratory conditions during this experiment, which may have added some extra constraints to the ones found in the field situation. The laboratory experiment represents a closed situation, while field conditions would be of the open system type. A long-term field experiment was therefore initiated.

Survival of microbes in long term buffer tests with in situ conditions

The long-term test (LOT) of buffer performance aims to study models and hypotheses of the physical properties of a bentonite buffer. Several species of bacteria with different relevant characteristics were introduced into the LOT bentonite. Mesophilic, thermophilic and spore-forming sulfate reducing bacteria, together with desiccation resistant, chemo-organotrophic or chemolithotrophic spore- and non-spore forming bacteria were mixed with bentonite clay to give approximately 100 million bacteria per gram (dry weight) of clay /Pedersen et al, 2000a/. The clay with bacteria was subsequently formed into cylindrical plugs with a 20 mm length and diameter, and installed in bentonite blocks exposed to low (20–30°C) and high (50–70°C) temperatures. The blocks were installed in the LOT boreholes immediately after the bacteria plugs were introduced /Pedersen et al, 2000a/. The experiment was terminated after 15 months. The major outcome was the effective elimination, to below detection limits, of all bacteria except the spore-forming ones. All of the three spore formers survived at the low temperature. The numbers remaining were, however, much lower than those initially introduced. The approximately 100 million spore-forming bacteria per gram (dry weight) of clay were reduced 100- to 10 000-fold. This can be interpreted as showing that the cell death rate was higher than the growth rate, which may have been zero, or close to zero. The spore-forming sulfate reducing bacterium was the only one of the three which survived at high temperature. It was concluded that the spore formers most probably survived as spores, and spores do not produce sulfide. Survival is not equivalent to activity. Since the methods used in this experiment did not reveal activity, additional experiments were set up to include measurement of sulfide production in simulated repository conditions.

Laboratory experiments exploring microbial sulphide production in buffer

The worst case scenario in copper canister corrosion would be if sulfate reducing bacteria formed biofilms on the canisters or grew intensively in the buffer close to the canister. To mimic this situation, several experiments have been executed and evaluated. In a first series, swelling pressure oedometers were loaded with bentonite at different densities, corresponding to different water activity values /Pedersen et al, 2000b/. A copper disc was placed between the bottom lid and compacted bentonite. Different sulfate reducing bacteria were added to the clay and the discs, together with $^{35}SO_4^{2-}$. The species used were laboratory cultures. Finally, oxidised silver foil was placed between the disc and the clay. The oedometers were reassembled and incubated for 4 weeks at the respective optimum temperatures and three different densities: 1500, 1800 and 2000 kg bentonite m^{-3}, corresponding to the water activities 0.999, 0.994, and 0.964, respectively. After incubation, $^{35}S\text{-}Ag_2S$ was localised on the silver foils and quantified by electronic autoradiographic imaging (Packard instant imager electronic autoradiography system, Meriden, U.S.A.). The amount of Ag_2S formed was used as a measure of the sulfate reducing activity.

The sulfate reducing bacteria used were active at a bentonite density of 1500 kg m^{-3}, but that sulfide production was virtually absent at higher densities. This experiment indicated that sulfate reducing bacteria probably cannot be active at the canister surface at a repository density of 2000 kg m^{-3}. However, the experiment was run with two laboratory species and it may be possible that other species which were not tested could survive better. Therefore, additional experiments was carried out using natural groundwater which commonly contains many hundreds of different microbial species, including several naturally occurring species of sulfate reducing bacteria.

Field experiments exploring microbial sulphide production in buffer

The activity of sulphate reducing bacteria (SRB) was again investigated in compacted bentonite with densities of 1500, 1800 and 2000 kg/m^3 using stainless steel oedometers with $^{35}SO_4^{2-}$ as the main sulphur source for the SRB /Masurat and Pedersen, 2003/. This time, the investigations were performed under in situ conditions, including a hydrostatic pressure of 25 bars at a depth of 450 m at the Äspö Hard Rock Laboratory. Canister type copper plates were placed in the oedometers, with the top either in contact with the added groundwater or with 3–4 mm bentonite between the plate and the added groundwater. The initial slot between bentonite and rock in a repository was simulated by a gap between the water inlet filter and the compacted bentonite. The bentonite density was thereby initially low at this end of the sample. The water uptake into the sample led to a density homogenization and a successive increase of density to the final test density. Radioactive hydrogen sulphide formed by the SRB reacted with the copper and formed radioactive copper sulphide, which was measured by electronic radiography as described for the lab experiments above. A similar set-up with filter-sterilized groundwater (0,2 µm) was used as control. The results showed that SRB were active and produced hydrogen sulphide during the initial phase of bentonite swelling. The activity of the SRB was inversely correlated with the final density of the bentonite as the highest density had the lowest overall SRB activity.

SRB presence in MX-80 bentonite

A second experiment was set up as the field experiment described above but adding heat treatment of the bentonite in the filtered groundwater control. The results showed that SRB were present in a dormant state in the commercial MX-80 bentonite. By addition of water, these dormant SRB became active and started to produce hydrogen sulphide. It is obvious that the sources of SRB to buffer material are two; the groundwater and the bentonite itself.

Natural analogues/observations in nature

There is a natural analogue in Dunarobba, Italy, where a bentonite slide buried a forest approximately one and a half million years ago. Trees that have not been decomposed by microorganisms can still be found. This indicates that conditions for microbial activity in bentonite clay are unfavourable.

Time perspective

The time perspective concerning survival and activity of microbes in compacted bentonite can be summarised as follows. At the time of deposition, there will be a canister, bentonite blocks and a hole in the rock. The next step will be to allow water to fill up all the void volume. This water can be groundwater from the rock or, alternatively, groundwater or water added from above at deposition. Irrespective of the source, microbes will be present in the water and these microbes will mix with the buffer, as described above.

The swelling of the clay will seed groundwater microbes into the clay, to a depth which is dependant on the initial slot, on both the rock and the canister side of the bentonite, and the microbes indigenous to the bentonite will be present in the entire bentonite buffer. The experimental data on survival and activity of microbes in bentonite suggest that the number of viable microbes will decrease rapidly during swelling and that very few viable cells will be present at full compaction. Sulfate reducing activity will also approach zero when full compaction is achieved and the only survivors will be microbes that have formed spores. Our results indicate that viable cell activity will be impossible at full compaction, as spores are inactive. Although spores generally are very resistant to difficult environmental conditions, they do still die off. All our experiments so far indicate a decrease in the number of viable spores at full compaction so that a slow but significant death rate of spores would eventually lead to the complete eradication of life in the buffer. It has not yet been clarified whether this will occur in the lifetime of a radioactive repository. Once the bentonite becomes sterile, it will probably not be reinfected. The theoretical pore size of the clay is 100–1000 times smaller than the average-sized microbe, meaning that no new microbes can enter into the buffer.

Handling in the safety assessment SR-Can

Microbial sulphide generating processes can be neglected for buffer densities at or above 1800 kg/m^3. This value is based on the fact that no microbial activity has been found in any of the performed experiments in which the bentonite density has been 1800 kg/m^3 or above during the whole course of the test. If the density decreases below this value, microbial processes will be inversely correlated with the density. Decreasing density will result in increasing microbial activity, including sulphide and potential gas production.

Uncertainties

Uncertainties in mechanistic understanding

The mechanisms for survival of microbes in bentonite and buffer materials are not fully understood. Evidence for the presence of viable microbes in commercial MX-80 bentonite suggests that SRB and also other species can withstand the very dry conditions (10%) in commercial bentonite. The present hypothesis is that microbes are introduced to the clay during mining and processing. The clay causes dehydration, which will gently remove water from the cell. The cells will be inactive, but viable. This process is similar to freeze drying of cells, which also results in inactive but viable cells, e.g. freeze dried bakers yeast. Adding water revives the microbes to an active life.

Commercial MX-80 is not compacted. The disappearance of microbial activity and viability in highly compacted bentonite is hypothesised to be a combined effect from desiccation and mechanical pressure. The sum of stress factors, high temperature, low water availability and a high mechanical pressure results in death of the microbes. However, this remains to be conclusively demonstrated.

Furthermore, the potential effect of initial bacterial activity during water saturation, and the exact critical bentonite density which leads to shut-off of bacterial activity remain to be determined.

The role of microbes in the redox processes has not been fully understood. This is of particular importance for the stability of the copper canister which could be hampered by sulphate-reducing bacteria. There are recent indications from the FEBEX experiments that these bacteria could be viable under compacted bentonite conditions, contrary to previous assumptions.

Model simplification uncertainties

The cut off boundary for microbial processes have been set to a buffer density of 1800 kg/m^3. This is a density where the experimental methods no longer observe microbial activity or prolonged survival. The possibility of some remaining activity, below the detection limits of current methods can not be rejected. The very long time of operation would then allow for some sulphide and gas production, but the rate would be very low.

Input data and data uncertainties

Not relevant since no model is used.

2.5.14 Speciation of radionuclides

Overview/general description

Speciation of radionuclides is of importance for sorption and diffusion in the buffer. It is influenced by what speciation the nuclide had at the boundary to the buffer, i.e. inside the canister, but also by the chemical conditions in the buffer.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	Possibly	The effect from radiolysis in the buffer will be small
Temperature	No	Yes	
Water content	No	No	
Gas content	No	No	
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	No	No	
Pore geometry	No	No	
Stress state	No	No	

Variable	Influencing?	Influenced?	Notes
Montmorillonite composition	(Yes)	Yes	Sorbed radionuclides will change the smectite composition, but the effect is limited
Pore water composition	Yes	Yes	
Bentonite composition	No	Yes	Indirectly through Pore water composition
Structural and stray materials	No	Yes	Indirectly through Pore water composition

Boundary conditions

The boundary condition for the process in the source term from the canister/fuel, ie the speciation inside the canister.

Model studies/experimental studies

If the water chemistry in the buffer is known, the speciation of radionuclides can be calculated with a thermodynamic equilibrium model, like PHEEQC or EQ3/6.

Time perspective

All.

Natural analogues/observations in nature

Not applicable.

Handling in the safety assessment SR-Can

The solubility and speciation of radionuclides is calculated and used as a source term for the modelling of release from a defective canister (See the forthcoming documentation of fuel processes). This is done for a wide span of water compositions.

In the buffer, the speciation is used to determine the transport properties of the radionuclides. However, the parameters for the transport properties used in SR-Can are selected based on experiments done under relevant conditions or from thermodynamic sorption models. See section 2.7.5 and 2.7.3. The consistency of these speciations with the speciation calculated in the source term will be checked.

Uncertainties

Uncertainties in mechanistic understanding

See processes section 2.7.5 and 2.7.3.

Model simplification uncertainties for the above handling in SR-Can

See processes section 2.7.5 and 2.7.3.

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

See processes section 2.7.5 and 2.7.3.

2.6 Radionuclide transport processes

2.6.1 Transport of radionuclides in water phase

Overview/general description

A number of processes will or could influence the transport of radionuclides in water phase: advection, diffusion, sorption, speciation, colloid transport and radioactive decay. All these have been discussed among the processes occurring in the buffer above, or, in the case of decay, under fuel processes.

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	Not in the buffer
Temperature	No	Yes	
Water content	No	(No)	Radionuclide transport is assumed to take place when the buffer is saturated
Gas content	No	(No)	Radionuclide transport is assumed to take place when the buffer is saturated
Hydrovariables (pressure and flows)	No	Yes	
Buffer geometry	No	Yes	
Pore geometry	No	Yes	
Swelling pressure	No	No	
Smectite composition	(Yes)	Yes	
Pore water composition	Yes	Yes	
Smectite content	No	Yes	
Impurity content	No	(No)	May have an effect under some circumstances
Structural and stray materials	No	No	

Boundary conditions

The inner boundary is determined by the size and shape of the defect in the copper canister and the concentrations of radionuclides in the water phase inside the canister.

The outer boundary is determined by the hydraulic properties of the rock surrounding the deposition hole, in particular fracture geometries and the Darcy velocity of the groundwater.

Model studies/experimental studies

No integrated experiments with radionuclide transport from a damaged canister, through a buffer into fractures in the rock have been performed. However, the individual processes governing the transport have been studied in detail. The most relevant processes in the buffer are 2.7.5, 2.7.3, 2.7.2and 2.7.4.

Natural analogues/observations in nature

Not applicable.

Time perspective

All timescales after canister failure.

Handling in the safety assessment SR-Can

Radionuclide transport in the buffer is handled in the integrated modelling of radionuclide transport for the repository. The near field code COMPULINK/COMP23 /Romero et al, 1999/ will be used for the calculations. The code calculates the non-stationary nuclide transport in the near field of a repository. The system is divided into compartments, where the only restriction is that a compartment is formed of the same material. The model, which is basically a coarsely discretised Integrated Finite Difference Model, embeds analytical solutions at locations which otherwise would require a very fine discretisation, such as entrances and exits from small holes and fractures. In the repository, radionuclides leaking out through a small hole in the canister wall diffuse into the buffer and then may migrate through various pathways into the flowing water in the rock fractures.

The buffer processes that need to be considered are treated in the following way:

- **Advection** is neglected since the buffer conductivity is < 10^{-12} m/s, see 2.5.2.
- **Diffusion** is treated with element specific diffusivities and porosities, see 2.7.3.
- **Sorption** is treated with linear sorption coefficients (Kd) for all elements, see 2.7.5.
- **Colloid transport** is neglected in the buffer, provided the buffer completely envelops the canister and has a dry density of at least 1,000 kg/m^3, i.e. a clay density of at least 1650 kg/m^3, see 2.7.4.
- **Speciation of radionuclides** is directly included in the calculation of the source term from the inner boundary condition. The speciation is also included in the determination of the sorption coefficient.
- **Radioactive decay** and chain-decay is considered in the calculations.

Radionuclide transport in the buffer is strongly dependent on the conditions at the inner and outer boundaries. The geometry of defects and fractures will determine the rate of transport.

An earthquake of large magnitude may potentially change the boundary conditions for the process, while the radionuclide transport within the buffer will remain the same.

A simplified, analytic model /Hedin, 2002/ will be used for scoping calculations of radionuclide transport in the near-field. This model is built on the same conceptual understanding of radionuclide transport in the buffer as COMP23.

Uncertainties

Uncertainties in mechanistic understanding

This process is an integration of several other processes all of which have uncertainties (see above).

Model simplification uncertainties for the above handling in SR-Can

The COMPULINK/COMP23 code uses rather a coarse discretisation of the buffer and analytical solutions for the boundary conditions at the canister and the rock.

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

The data uncertainties for the transport parameters in the buffer are presented in process 2.7.5 and 2.7.3. All input data to the radionuclide transport calculations will be handled in the SR-Can data report.

2.6.2 Transport of radionuclides in gas phase

A pulse of hydrogen gas can be released from a defective canister see section 2.5.3. Some radionuclides could potentially enter the gas phase and thereby be transported to the surface much more rapidly than would be the case for the aqueous pathway. In principle, only C-14 and Rn-222 can enter the gas phase.

Overview/general description

The following table summarises how the process influences and is influenced by all buffer variables.

Variable	Influencing?	Influenced?	Notes
Radiation intensity	No	No	
Temperature	No	No	The thermal phase will be over before high enough gas pressures can be reached
Water content	No	No	
Gas content	No	(No)	The process is assumed to occur only when the buffer is saturated
Hydrovariables (pressure and flows)	No	No	
Buffer geometry	No	(Yes)	The opening pressure may be dependent on the geometry (see section 2.5.3)
Pore geometry	No	No	
Swelling pressure	No	Yes	The opening pressure is dependent on the swelling pressure(see 2.5.3)
Smectite composition	No	(Yes)	Indirectly through swelling pressure
Pore water composition	No	(Yes)	Indirectly through swelling pressure
Smectite content	No	(Yes)	Indirectly through swelling pressure
Impurity content	No	No	
Structural and stray materials	No	No	

Boundary conditions

The inner boundary for this process is the gas pressure within a defective canister. The outer boundary is the fractures in the wall of the deposition hole.

Model studies/experimental studies

Not done for relevant conditions

Natural analogues/observations in nature

Not relevant.

Time perspective

The process will only occur if there is a defect in a canister and if a sufficient amount of water is supplied to the iron insert to generate, over time, a gas pressure that is higher than the opening pressure of the bentonite.

Handling in the safety assessment SR-Can

Radionuclide transport in a gas phase will be treated as a separate case in SR-Can. The handling is based on the experiment by /Harrington and Horseman, 2003/ presented in section 2.5.3. It is assumed that the bentonite will open and release gas when the internal pressure exceeds 20 MPa. A rapid outflow will occur until the pressure falls to 10 MPa. This means that half of the gas inside the canister will be released instantaneously together with the radionuclides contained in that amount of gas. Neither the buffer nor the geosphere is expected to delay the transport to the biosphere. After the breakthrough pulse the gas is expected to be released at the same rate as it is produced by corrosion.

Uncertainties

Uncertainties in mechanistic understanding

The mechanisms for gas release through bentonite are still not fully understood, see section 2.5.3.

Model simplification uncertainties for the above handling in SR-Can

The handling of the gas transport pathway in SR-Can is based only on a few experiments. The values for opening and steady state pressures have been selected on from those experiments.

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

As mentioned above, the values for gas opening and state pressures are based on experimental evidence. However, under any conditions, the gas pressure differential should be in the range of 5–15 MPa. Another uncertainty is the fraction of C-14 that can enter a gas phase. In the safety assessment SR 95 /SKB, 1995/, the value of 3.5% was selected based on /Codell, 1993/. This value may be associated with a large uncertainty. For Rn-222, it is probably fair to assume that all of the inventory will be in the gas phase.

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Appendix A

A model describing buffer erosion

Introduction

In the KBS 3 concept for a final repository for spent nuclear fuel the canisters containing the spent are placed in large drilled holes in the floor of tunnels in crystalline rock at about 500 m depth. The buffer around the copper canisters consists of compacted bentonite clay. The clay, when wetted by water swells strongly, fills out any voids in the deposition hole and protects the canister from flowing water and mechanical movement of the rock. Any clay that may have expanded out into a fracture intersecting the deposition hole will not be swept away by the seeping water because the clay particles are kept together and form a gel. However, should the water that normally is slightly salty in deep crystalline rocks be intruded by fresh water, the gel can disperse and the small clay particles could be swept away as colloids.

This could lead to loss of buffer. The colloidal particles could also form carriers for any escaping nuclides, which could be carried by the colloids with the seeping water.

This note sets out to explore the mechanisms that could lead to clay dispersal and to attempt to devise a model to quantify the rate of dispersal.

Bentonite

The bentonite clay that will be used contains a high proportion of montmorillonite, more than 80%. The cation exchange capacity, CEC, is on the order of 0.75 meq/g. The cations are mostly Na and Ca but there is also some Mg and K.

The table is taken from SR Can Interim report for SIERG comments

Table A-1. Impurities and accessory minerals in MX-80 and Deponit CA-N bentonite.

Component	MX-80 (wt-%)	Deponit CA-N (wt-%)	Uncertainty (± wt-%)
Calcite + Siderite	0	10	1
Quartz	3	1	0.5
Cristobalite	2	1	0.5
Pyrite	0.07	0.5	0.05
Mica	4	0	1
Gypsum	0.7	1.8 (anhydrite)	0.2
Albite	3	0	1
Dolomite	0	3	1
Montmorillonite	87	81	3
Na–	72%	24%	5
Ca–	18%	46%	5
Mg–	8%	29%	5
K–	2%	2%	1
Anorthoclase	0	2	1
CEC (meq/100 g)	75	70	2

The montmorillonite particles are made up thin sheets of one layer of aluminium oxide sandwiched between two silicon oxide layers. The edges have different properties than the flat sides. The particles have a net negative charge due to isomorphic substitution where a silicon atom in the SiO2 structure has been exchanged for an aluminium atom. This negative charge is compensated by the presence of a loosely bound cation, often Na and Ca. The oxides may hydrolyse when in contact with water and form surface bound hydroxyl groups, =S-OH groups. =S stands for surface. The proton on the =S-OH group may detach at higher pH or be exchanged for a cat-ion to for =S-O$^-$ or =S-O$^-$Na$^+$. /Stumm and Morgan, 1996/.

The cations that are attracted mainly by the negative charge of the particle as well as the surface complexes play a major role for the stability of colloidal gels.

The table below is directly copied from /Bruno et al, 1999/.

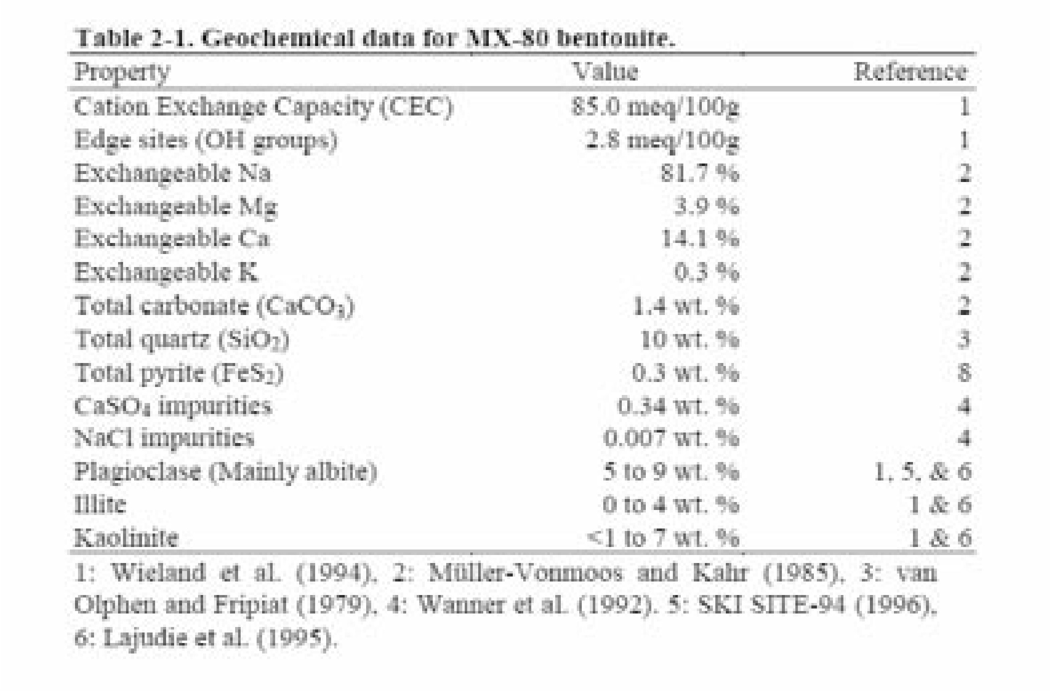

Table 2-1. Geochemical data for MX-80 bentonite.

Property	Value	Reference
Cation Exchange Capacity (CEC)	85.0 meq/100g	1
Edge sites (OH groups)	2.8 meq/100g	1
Exchangeable Na	81.7 %	2
Exchangeable Mg	3.9 %	2
Exchangeable Ca	14.1 %	2
Exchangeable K	0.3 %	2
Total carbonate ($CaCO_3$)	1.4 wt. %	2
Total quartz (SiO_2)	10 wt. %	3
Total pyrite (FeS_2)	0.3 wt. %	8
$CaSO_4$ impurities	0.34 wt. %	4
NaCl impurities	0.007 wt. %	4
Plagioclase (Mainly albite)	5 to 9 wt. %	1, 5, & 6
Illite	0 to 4 wt. %	1 & 6
Kaolinite	<1 to 7 wt. %	1 & 6

1: Wieland et al. (1994), 2: Müller-Vonmoos and Kahr (1985), 3: van Olphen and Fripiat (1979), 4: Wanner et al. (1992), 5: SKI SITE-94 (1996), 6: Lajudie et al. (1995).

Ground water compositions

The groundwaters in deep Swedish crystalline rocks contain dissolved salts. Na$^+$ and Ca^{2+} are commonly the main cations but there also is Mg^{2+} and K$^+$ and various other minor constituents. Cl$^-$, HCO3$^-$ and SO4^{2-} are the most abundant anions. Typically the Ca^{2+} and Mg^{2+} have concentrations near or above 1 mmol/l. This concentration of divalent ions is around the CCC, the Critical Coagulation Concentration for the montmorillonite particles. At higher groundwater concentrations the particles will form a stable gel, at much lower concentrations the gel will break up and the particles disperse. They will then not attach to each other or to other aluminosilicate minerals that make up the crystalline rock. The particles will therefore be carried by the water and could bring with them any adsorbed radionuclides.

The table below is directly copied from /Bruno et al, 1999/.

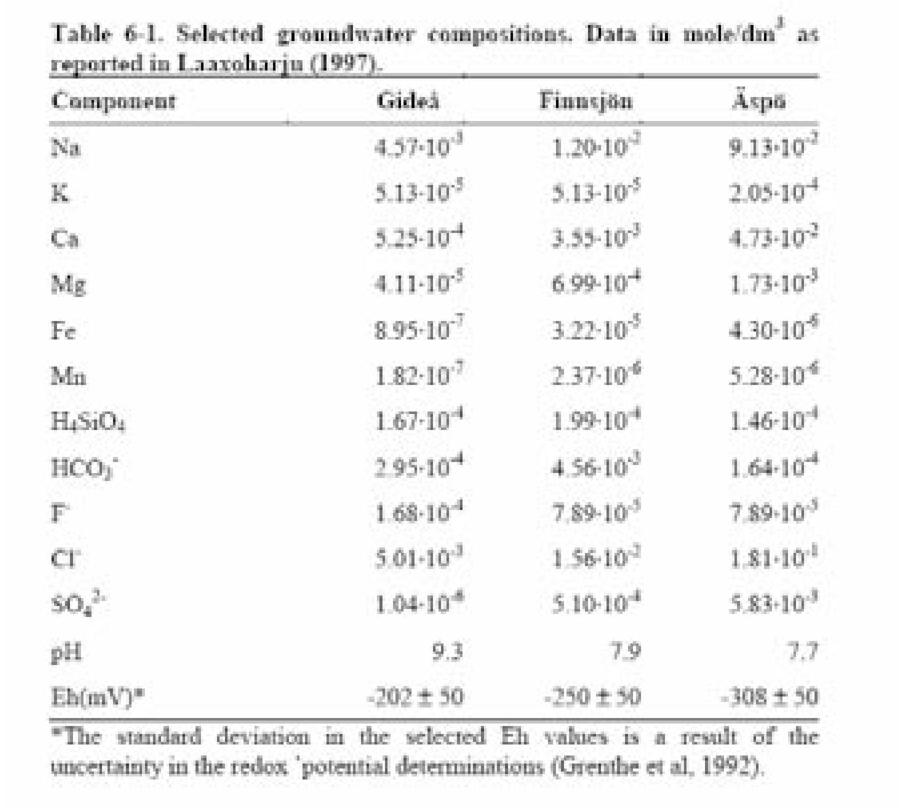

Table 6-1. Selected groundwater compositions. Data in mole/dm^3 as reported in Laaxoharju (1997).

Component	Gideå	Finnsjön	Äspö
Na	$4.57 \cdot 10^{-3}$	$1.20 \cdot 10^{-2}$	$9.13 \cdot 10^{-2}$
K	$5.13 \cdot 10^{-5}$	$5.13 \cdot 10^{-5}$	$2.05 \cdot 10^{-4}$
Ca	$5.25 \cdot 10^{-4}$	$3.55 \cdot 10^{-3}$	$4.73 \cdot 10^{-2}$
Mg	$4.11 \cdot 10^{-5}$	$6.99 \cdot 10^{-4}$	$1.73 \cdot 10^{-3}$
Fe	$8.95 \cdot 10^{-7}$	$3.22 \cdot 10^{-5}$	$4.30 \cdot 10^{-6}$
Mn	$1.82 \cdot 10^{-7}$	$2.37 \cdot 10^{-6}$	$5.28 \cdot 10^{-6}$
H_4SiO_4	$1.67 \cdot 10^{-4}$	$1.99 \cdot 10^{-4}$	$1.46 \cdot 10^{-4}$
HCO_3^-	$2.95 \cdot 10^{-4}$	$4.56 \cdot 10^{-3}$	$1.64 \cdot 10^{-4}$
F^-	$1.68 \cdot 10^{-4}$	$7.89 \cdot 10^{-5}$	$7.89 \cdot 10^{-5}$
Cl^-	$5.01 \cdot 10^{-3}$	$1.56 \cdot 10^{-2}$	$1.81 \cdot 10^{-1}$
SO_4^{2-}	$1.04 \cdot 10^{-6}$	$5.10 \cdot 10^{-4}$	$5.83 \cdot 10^{-3}$
pH	9.3	7.9	7.7
Eh(mV)*	-202 ± 50	-250 ± 50	-308 ± 50

*The standard deviation in the selected Eh values is a result of the uncertainty in the redox 'potential determinations (Grenthe et al, 1992).

Colloidal properties

A negatively charged surface in water will attract cations. Some cations may be directly bound as surface complexes $=S\text{-}O^-Me^+$. The negative $=SO^-$ charge is then neutralized by the specifically bound metal, Me^+. However, there remain those sites that have not formed surface complexes and all the fixed charges. These are balanced by a surplus of cations present in what is called a diffuse layer very near the surface. This layer also has a lower concentration of anions as these are repelled by the negatively charged surface. At equilibrium the net surplus of positive charges in the diffuse layer exactly compensate the surface charge. Obviously, the higher total concentration there is in the water the thinner the diffuse layer need to be to be able to contain the charges needed for the balance. Also divalent or even higher valence ions cations are more effective to balance the negative charge of the surface.

The pH of the water will also influence these processes because the surface sites will tend to be more protonated at low pH than at high pH. Thus the pH, the total concentration of dissolved ions and the charge of the cations will influence the thickness of the diffuse layer.

This implies that a high salt concentration will tend to cause a thin diffuse layer and therefore the particles can be located nearer to each other without the diffuse layers of two particles meeting. The repulsive forces between particles will then be small at a given distance. The contrary will be true in low salinity waters. Then the diffuse layers extend far from the surface and the particles will repel each other. This is the reason why the backfill has a higher swelling pressure in low salinity waters than in high. It also explains why a Ca-bentonite has less swelling pressure than a Na-bentonite.

These phenomena also suggest that a low salinity water will tend to disperse the particles more than a high salinity water. In the latter case when the particles are close together without there being strong repulsive forces, other attractive forces can dominate and lead to the formation of stable gels.

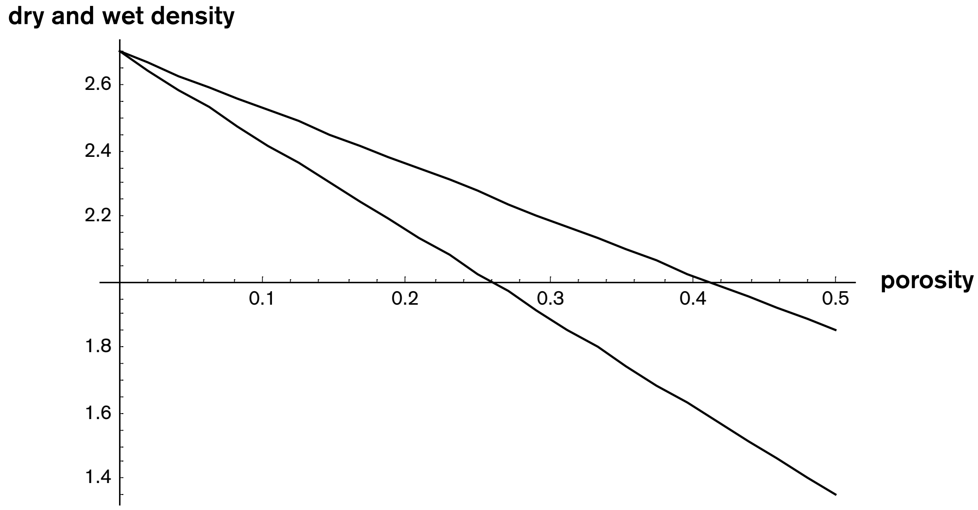

Figure A-1. *Wet (upper curve) and dry (lower) density of compacted bentonite as a function of porosity.*

In order to assess whether a bentonite will for a gel or will disperse as a sol the CCC of the bentonite as well as the groundwater composition must be known /Evans and Wennerström, 1999/.

The CEC of the bentonite is around 1mmol/l of divalent ions that is the sum of Ca^{2+} and Mg^{2+} in the expected waters. /LeBel, 1978/.

The porosity of the compacted bentonite is on the order of 30–50%, for wet densities of the bentonite between 1800 and 2200 kg/m^3.

Processes

At steady state the pore water of the bentonite in the deposition hole is in equilibrium with the surrounding water. The water composition in the pores of the compacted bentonite is not the same as that of the water outside because the presence of the surfaces of the bentonite influences the pore water composition. The difference is larger the more compact the bentonite is. However, at first we neglect this difference because the emphasis of this note is on conditions where the clay has expanded considerably and is on the verge of dispersing. Then the error introduced in neglecting the influence of the clay will not be large in comparison to some of the other uncertainties such as CCC, actual clay and water composition etc.

Thus assume that the CCC is known and that that when the water composition in the clay in the vicinity of the clay water interface should become less than the CCC the clay will disperse.

The task is then to assess how the pore water of the expanding clay in a fracture will change with time and how much clay will be influenced and dispersed.

Model and sample calculations

The following conceptualisations are used to set up the model.

The clay in the deposition hole has a swelling pressure that acts as a force to push out the clay into the fracture. As the clay moves out into the fracture it expands and the swelling pressure decreases with decreasing clay density. During swelling the clay takes up water and equilibrates with the local water composition. There will therefore be a gradient of water content as well as of the water chemistry along the swelled clay. A final state is eventually approached when the friction force of the clay against the walls of the fracture balances the force that pushes the clay out into the fracture. At the outer rim of the clay the clay forms a stable gel and no particles are carried away if the salt concentration is above the CCC.

Next, let the water composition at the outer edge of the clay change so that it will be lower than the CCC. For simplicity of description assume that it is the calcium concentration in the water that determines the CCC. Also consider a case where there is a constant exchange of water with a known rate Q_{eq} with an approaching concentration c_w. When the pore water has a higher concentration of calcium than the water outside the clay, calcium diffuses out from the gel to the water. The water will take up calcium and leave with a concentration c_i.

At some point in the gel the calcium concentration would become less than CCC. The clay particles will disperse and be carried away by the water. This implies that the concentration at the gel/sol interface will be equal to the CCC. This will also be the concentration in the leaving water. Thus c_i= CCC.

This of course is a simplified model because in the fracture the approaching water will first meet clay that gives off calcium, which has to diffuse out further in the water in the fracture. There will therefore develop a concentration gradient in the water that passes the clay and the longer the water has been in contact with clay the further out the calcium has diffused. This can be handled by the boundary layer theory and has previously been used to describe the transport to and from the clay at the fracture interface. On the average the transport capacity of the flowing water can be summarized in an "Equivalent Flowrate" Q_{eq} (m^3/yr). This is the flowrate of water that will carry away the calcium with a concentration equal to that at the interface, ci minus that in the approaching water c_w. The calculations could be refined to account for the changing conditions as the water passes the clay interface but that will not be done in the present note.

In this first simplified model, assume that a steady state will prevail so that a constant clay distance and density gradient is established. Assume further that the supply of clay is unlimited from the deposition hole.

Figure A-2 illustrates the major processes; clay expansion, clay dispersion, dissolution of mineral and diffusion of dissolved Ca towards the water. In the simplified model the dissolution rate is assumed constant, the diffusion resistance in the clay is neglected and the diffusion resistance in the seeping water is averaged and summarized in the concept of equivalent flowrate, Q_{eq}.

A mass balance can now be made of how much calcium, m_{Ca}, (mol) is carried away during a given time. It is

$$m_{Ca} = Q_{eq} * (c_i - c_w) * time \qquad (1)$$

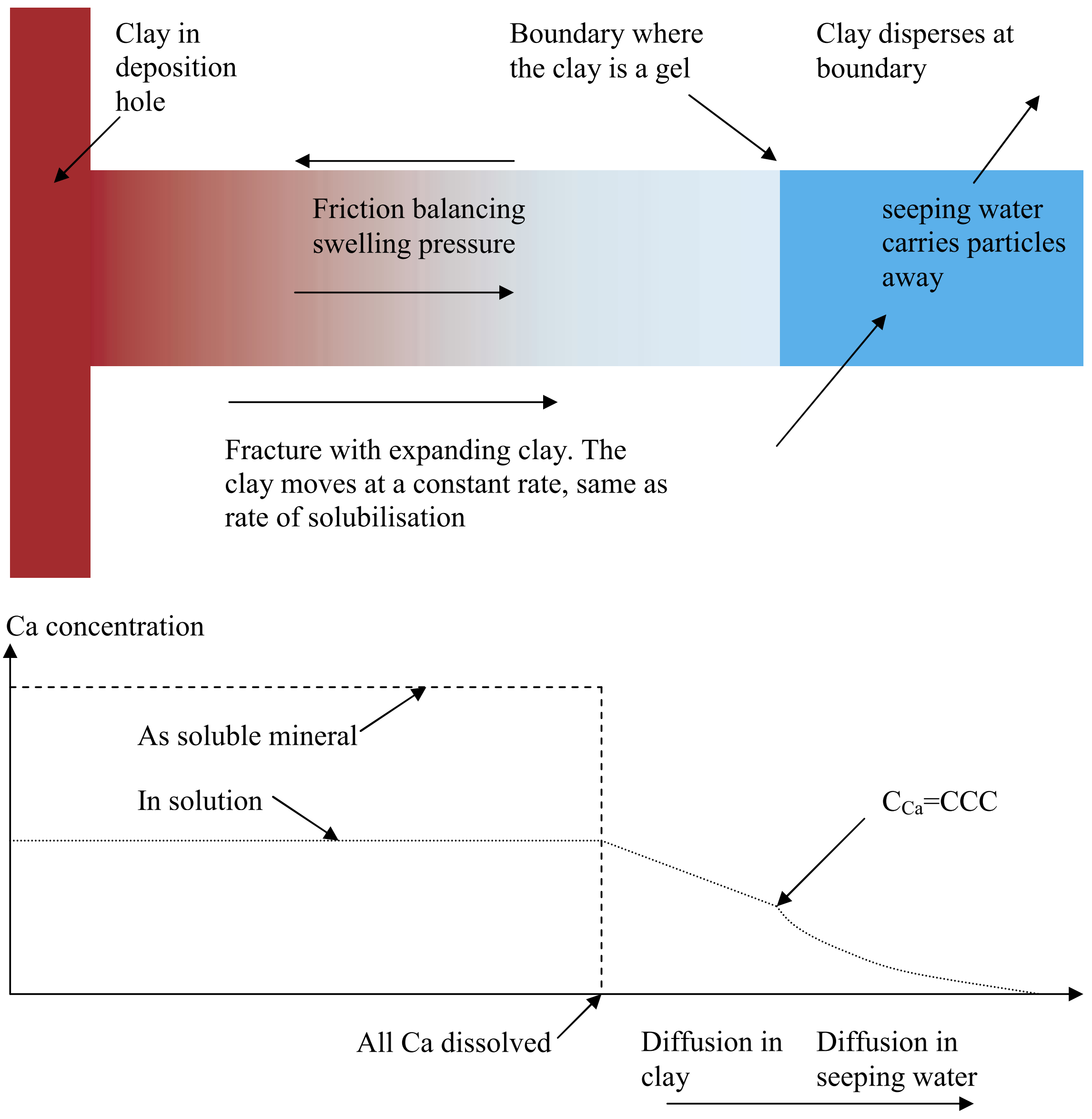

Figure A-2. *Clay swells into a fracture. Ca minerals dissolve and Ca diffuses towards the seeping water. Ca concentration drops to CCC. Clay disperses into the water and is carried away.*

That calcium must have come from the pore water of the original clay. The volume of "original" clay that enters the fracture carries with it the calcium in its pore water as well as any soluble Ca minerals. The clay will be depleted of calcium from the original concentration c_o to that at the interface c_i. c_o is the concentration of any soluble calcium mineral and of Ca in the original pore water outside the diffuse electrical layer. Also the Ca bound as surface complex is not included[1]. Thus m_{Ca} is also obtained from the mass balance for the calcium in the clay pore water and ε_{clay} the porosity of the clay.

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[1] The influence of Ca in the diffuse double layer is that which is bound as cation exchangeable species. The interaction with the ion exchange is neglected in this simplified model. It will have to be considered later. The error introduced by this is not negligible because the CEC is on the order of 0.75 mmol/g of which 17% is CaX_2. There is thus 0.08 mmol/g Ca bound as Ca X_2. The Ca mineral content of the clay is on the order of 1% (MX-80) giving about 10 mg Ca mineral per gram clay, which in turn is about 0.1 mmol/g. In a full model both forms of Ca must be accounted for.

$m_{Ca} = V_{clay} * \varepsilon_{clay} * (c_o - c_i)$. (2)

Equating Equations (1) and (2) gives the clay volume V_{clay} depleted of calcium. This volume contained a mass of clay m_{clay}

$m_{clay} = \rho_{dry,clay} * V_{clay}$ (3)

Then the loss of clay per unit time N_{clay} (kg/yr)

$N_{clay} = Q_{eq} * (c_i - c_w) / \varepsilon_{clay} / (c_o - c_i) * \rho_{dry,clay}$ (4)

Example

The data in the table below are used to exemplify

Two cases are considered. In the first there are no calcite minerals. In the second there is 1% by weight of calcium ($x_{CaSolid}$ =10g Ca/kg solid clay-> 0.25 mol Ca /kg clay). The latter gives

$c_{o,in\ solids} = x_{CaSolid} / M_{Ca} * \rho_{clay} / \varepsilon_{clay}$ (5)

Entity	unit	
Qeq (m^3/yr).	(m^3 water /yr).	0.001
co	mol/m^3 pore water	10 (No Ca mineral) , 1250
CCC=ci	mol/m^3 pore water	1
cw	mol/m^3 pore water	0
ρdry,clay	kg/m^3 dry clay	1600
εclay	m^3 pore water/m^3 wet clay	0.4

In the first case 0.4 kg of clay will be carried away per year, in the second case 3.2 g year will be carried away.

In 1000 years it will be 400 kg and 3.2 kg respectively.

Discussion and conclusions

This very simple approach suggests that if there are soluble Ca or Mg minerals with on the order of percent by weight in the clay these will be able to supply the clay pore water with divalent ions and to a large extent prevent the clay from dispersing in the passing water in the fracture.

However it should be noted that a number of simplifying assumptions have been used in this first scoping model.

The following assumptions must be revisited and checked for reasonableness

1. The change of composition in the diffuse layer is not accounted for
2. Steady state assumption for clay intrusion into the fracture
3. Steady state assumption of the chemical transport

4. Applicability of Q_{eq} model
5. Assumption that the Ca transport in the bulk of the clay in the deposition hole can be neglected
6. Assumption of no mechanical erosion of gel fringe
7. Assumption of Ca mineral dissolution
8. Assumption of steady state for chemistry- e.g. no pH changes that influence the CCC.

The above items can all conceivably influence (increase) the dispersion rate. There may be more that will be found in a more detailed investigation

The following could act to decrease the dispersion rate

1. The diffusion resistance in the clay in the fracture is neglected
2. The change of composition in the diffuse layer is not accounted for
3. Physical hindering of clay particle dispersion in the fracture
4. Clogging of water pathways by the clay being pressed out into the fracture.
5. Sedimentation of clay particles in fracture
6. Development of a montmorillonite depleted region near the fracture entrance leaving behind sparsely soluble minerals that “clog” the fracture entrance

There may be more effects that will be found in a more detailed investigation