BELBaR D1.2

STATE-OF-THE-ART REPORT ON THE TREATMENT OF COLLOIDS AND RELATED ISSUES IN THE LONG TERM SAFETY CASE

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Foreword

This report, a key early-project deliverable for the EC BELBaR project, considers the current state-of-the-art treatment of bentonite colloid issues in performance assessment (PA) for a range of participating national radioactive waste management programmes.

The aims of the report are, for each waste management organisation represented in the BELBaR project, to:

- Describe the disposal concept;
- Describe the current treatments of colloid issues in PA;
- Identify positives and negatives of the current treatment;
- Analyse limitation of previous studies and uncertainties related to colloids;
- Discuss needs for additional studies of colloid related issues, and PA relevance;
- Identify relevance of and expected benefit from BELBaR WPs 2-5 work.

Through the work being undertaken in the BELBaR project, progress will be made against the topics noted in this report. This report will be updated at the end of the BELBaR project to reflect this progress – the revised version of the report will clearly demonstrate how the BELBaR project has been of significant benefit to the national programmes that are participating in it with regard to the treatment of colloids in performance assessment.

Acknowledgements

This report is a product of Work Package 1 of the BELBaR project. All participating organisations and personnel are thanked for their significant input that determined its structure and contents, and for associated texts that collectively make this report.

1 Introduction

This report, a key early-project deliverable for the EC BELBaR project, considers the current state-of-the-art treatment of colloidal issues in performance assessment (PA) for a range of participating national radioactive waste management programmes.

The aims of the report are, for each national programme, to:

- Describe the disposal concept;
- Describe the current treatments of colloid issues in PA (on the basis of waste management organisations involved in BELBaR only);
- Identify positives and negatives of the current treatment;
- Analyse limitation of previous studies and uncertainties related to colloids;
- Discuss needs for additional studies of colloidal issues, and PA relevance;
- Identify relevance of and expected benefit from BELBaR WPs 2-5 work.

Section 2 of the report provides information on the structure of the BELBaR project, noting how the five constituent Work Packages are inter-related and designed to provide an information flow throughout the project. This relationship between the Work Packages is intended to ensure, for example, that the development of understanding of colloids processes at a research level is reflected appropriately in the treatment of colloids in performance assessment, and that the research studies themselves are focussed at the outset on key performance assessment issues – this is an iterative process, necessitating the close collaboration of a range of scientific disciplines and the undertaking of needs-directed research activities.

In **Section 3**, each country represented in BELBaR Work Package 1 has produced a description of how it currently considers colloids in performance assessment – the current state-of-the-art – following the above bullet points as a guide.

Information presented in Section 3 is subsequently used to identify current issues affecting the treatment of colloids in performance assessment in national programmes. These issues are identified in **Section 4** in a series of tables.

The issues are linked in these tables as appropriate to the Work Packages considered in the BELBaR project (which are described later in this section), clarifying how the issues raised by national programmes regarding the treatment of colloids in performance assessment will be progressed by the BELBaR project itself. Note that the relevance of the information contained in these tables to national programmes varies on a programme by programme basis.

Section 5 of this report presents a brief summary. **Section Fel! Hittar inte referenskälla.** is a glossary and **Section 6** presents a collation of references.

Through the work being undertaken in the BELBaR project, progress will be made against the issues noted in Section 4 of this report. This report will be updated at the end of the BELBaR project to reflect this progress – the revised version of the report will clearly demonstrate how the BELBaR project has been of significant benefit to the national programmes that are participating in it with regard to the treatment of colloids in performance assessment.

This report was finalised in October 2012 according to the schedule of the EC BELBaR project. It meets BELBaR deliverable 1.1. It presents the country-by-country current state-of-the-art regarding the treatment of colloid issues in performance assessment (PA) as at September 2012.

2 Background

The collaborative project is based on the desire to improve the long-term safety assessments for repository concepts that combine a clay EBS with a fractured rock. The formation and stability of colloids from the EBS may have a direct impact on assessed risk from the repository in two aspects:

- Generation of colloids may degrade the engineered barrier; and
- Colloid transport of radionuclides may reduce the efficiency of the natural barrier.

An increased understanding of processes will have an effect on the outcome of future assessments. Colloid related issues are relevant in the safety cases of nuclear waste disposal in crystalline rocks. More specifically, since natural colloids concentrations in typical repository conditions are negligible the most outstanding potential for colloids are in the colloids generated from clay and cementitious components.

These colloids are relevant from the point of view of probability of colloid mediated radionuclide transport and as a consequence of inappropriate degeneration of the engineered barrier system. Thus pre-eminent changes in the properties of the system include:

- decrease of clay density or the density of cementitious materials at and near their interface with groundwater with respect to the initial density causing further changes in some performance indicators such as the hydraulic conductivities of components in question;
- increase of the thickness of the diffusion barrier at the locations in which fractures intersect clay components;
- increase in hydraulic conductivity at the locations in which fractures intersect cementitious components; and
- increase in the amount of colloids available as carriers for radionuclides.

In this context the focus is on clay colloids and cement derived colloids are omitted from this project.

2.1 BELBaR Work Package Activities

In the following sub-sections, the structure and activities to be progressed in Work Packages 1-7 are briefly described.

2.1.1 WP1: Safety assessment (RWMD lead)

WP1 has the responsibility to ensure that that the type and values of the parameters selected for experimental and modelling work are those that will bring results corresponding as far as possible to all the different situations that can be expected in a geological disposal facility (GDF). WP1 has defined these parameters and values in an internal report at the beginning of the project [Ref?]. This information is being used to inform the experimental and modelling efforts in the other work packages.

At the start of the project, this work package will collect and present the current treatment of the relevant processes in safety assessments. This information will be used to the experimental and modelling efforts in the other work packages. Later, drawing on the work undertaken in WP 2 to 5, the general objective of WP1 is to consider how colloids and related phenomena can be considered in the long-term safety case, to 1 million years following the closure of a geological repository, and to make recommendations on the quantitative and qualitative approaches that a safety case could pursue to adequately address this potentially very significant issue.

2.1.2 WP2: Bentonite erosion (Ciemat lead)

The main objective of WP 2 will be to understand the main mechanisms of erosion of clay particles from the bentonite surface and to quantify the (maximum) extent of the possible erosion under different physico-chemical conditions.

Additionally, these studies will point out under what conditions compacted bentonite is able to produce colloidal particles, free to move into the contacting aqueous phase and to determine the bentonite colloids 'source term'. Whilst, the colloids-radionuclide and colloids-rock interactions, as well as effects of colloids on radionuclide transport will be analysed in WP3.

Data obtained at a laboratory scale and their interpretation, will be compared to those obtained in-situ at the FEBEX gallery at the Grimsel Test Site (GTS, Switzerland) where a real-scale experiment, simulating a HLW repository in granite, was installed 14 years ago. The compacted bentonite has been present in the FEBEX gallery for more than one decade, and colloid analysis to identify the presence of bentonite colloids in the groundwater near the bentonite surface has being carried out since 2006 (EC-FUNMIG Project).

The joint analysis of laboratory studies from WP2, WP3 and WP4 and in-situ data represents a good starting point for providing realistic inputs for the models used in the performance assessment (PA) of HLW repositories and their conceptualisation (WP1 and WP5).

2.1.3 WP3: Radionuclide and host rock interactions (KIT lead)

Clay colloids potentially generated in the radioactive waste repository near-field from the bentonite-buffer/backfill material (see WP2) might be stable under the geochemical conditions of the fractured rock far-field (see WP4) and could be a carrier of radionuclides. Colloid mobility is strongly dependant on fracture geometry (aperture size distribution and fracture surface roughness) as well as chemical heterogeneity induced by the different mineral phases present in the fracture filling material and the chemistry of the matrix porewater. This work package addresses the following topics:

1. The process understanding of colloid mobility controlling processes and their appropriate description. A bottom-up approach starting from mono-mineralic single crystals over fracture filling mineral assemblages to natural fractures will be used to identify the colloid attachment probability determining processes.
2. The mobility of clay colloids will not necessarily enhance the mobility of strong sorbing radionuclides, if the sorption is reversible. Strong radionuclide clay colloid sorption reversibility kinetics have frequently been observed, but the reasoning for the observed kinetics is still pending and detailed species determination is needed in order to implement these reactions in thermodynamic models.
3. Identifying additional retention processes. Colloid transport and naturally occurring colloid concentrations in fractured rocks are frequently correlated to the water chemistry found in the water conducting features. However, the potential release of divalent cations (Ca^{2+}, Mg^{2+}) via matrix diffusion is expected to increase the colloid attachment probability and reduce the colloid mobility even under glacial melt water/meteoric water conditions and has to be investigated.

2.1.4 WP4: Colloid stability (NRI lead)

Clay colloid stability is one of the key questions for prediction of their potential influence on erosion of bentonite buffer/backfill material (see WP2) and migration of radionuclide carried by colloids (see WP3). Clay colloid stability in aquatic environments is primarily driven by groundwater geochemistry such as salinity (ionic strength) and edge charge, which is a function of pH. Also other factors should be taken into account within the colloid stability such as water flow rate, filtration effects, heterogeneity of solid phase surface charge and time. The stability of clay colloids in the site-specific host rock conditions is important for assessments of long-term performance of radioactive waste repositories.

This work package focuses on:

1. Clay colloid stability studies under different geochemical conditions with respect to ionic strength and the pH. The colloids formed at the near/far field interface would be stable only if favourable conditions exist and therefore their relevance for radionuclide transport will be strongly site-specific.
2. Effects of removing colloidal particles from the liquid phase (such as reaching critical coagulation concentration, the effect of surfactants, coagulation).
3. Understanding the influence of complexing agents (organic / humic substances) on clay colloid stability to reduce the uncertainty of naturally occurring organic matter presence.

2.1.5 WP5: Conceptual and mathematical models (Posiva lead)

The objective of this work package is to validate and advance the conceptual and mathematical models used to predict mass loss of clay in dilute waters and clay colloid generation as well as clay colloid facilitated radionuclide transport relevant to geological disposal of higher level radioactive waste. Validation of the current conceptual as well as mathematical models is pending. The target is to obtain validated advanced model(s) for the purposes of geological disposal of higher level radioactive waste.

Modelling is a necessity to assess the processes relevant to geological disposal. This is because the related time scales are moderate in geological sense but far too long in terms of experimental assessments regarding the time scales of industrial product development. Models can be validated with small-scale experiments provided that scaling can be reasoned using discipline wise accepted understanding of processes and conditions relevant to the application.

Validation with the data obtained from WP2-4 will be used to develop conceptual understanding, the related mathematical models and to provide feedback to numerical implementation of the models. The overall understanding will later be used in the safety assessment formulations in WP1.

In validation at least the following aspects will be weighed up:

1. Preparedness to predict the data obtained from WP2-4 and other possible test cases, e.g. by judging:
    a. whether the values of the targeted indicators are predicted to be at the right order of magnitude and
    b. whether the major changes in slopes (cf. inflection points) in terms of dominating parameters occur at the right order of magnitude,
2. Adequacy of the reasoning for the selection of the processes considered as dominant, and
3. Identification and reasoning of the parameters of relevance.

The results of the studies will also be used as aids for the design of new/additional experiments in WP2-4.

2.1.6 WP6: Knowledge management, dissemination and training (SKB lead)

To ensure that BELBaR has the appropriate impacts, a specific work package (WP6) has been established for dissemination. It is anticipated that a range of tools will be used to reach a wide audience. In particular, it is thought that the results will be disseminated to international conferences, high level publications including a special issue of a journal or published proceedings from a workshop, engaging with 3rd party organisations, and training at a summer school or professional development courses. The work package leader of WP6 will also act as a cross-cutting theme coordinator.

BELBaR will use a number of avenues to disseminate knowledge with the project website providing the primary initial interface for partners and the wider scientific community. Multiple tools covering a wide range of formalities will be used (e.g. website, project meetings, reports, workshops, journal publications, conferences).

2.1.7 WP7: Co-ordination (SKB lead)

WP7 will provide adequate administrative, legal and financial management of the project, and documentation of scientific-technical progress through Annual Workshop Proceedings.

The Project Coordinator, Legal Officer and Financial Officer, will take all actions required in order to ensure:

- Overall functioning of the project work and activities,
- Communication between the project and the Commission, including submission of the required management and activity reports,
- Monitoring of the use of resources and transferring financial resources,
- Ensure Annual Project Workshops and organize project meetings,
- Organize and publish Annual Workshop proceedings, and
- Take any other action required in support of optimized project implementation.

3 Current Treatment of Colloids in Performance Assessment

In this section, each waste management organisation (WMO) represented in BELBaR Work Package 1 has produced a description of how it currently considers colloids in performance assessments. The format used to guide these sub-sections is:

- Describe disposal concept;
- Describe the current treatment of colloid issues in PA (on the basis of organisations involved in BELBaR only);
- Identify positives and negatives of the current treatment;
- Analyse limitation of previous studies and uncertainties related to colloid transport;
- Discuss needs for additional studies of colloid issues, and PA relevance.

A series of tables is presented, in which the issues are considered and linked as appropriate to the Work Packages considered in the BELBaR project (see Section 1 for further details), clarifying how the issues raised by national programmes regarding the treatment of colloids in performance assessment will be progressed by the BELBaR project itself.

Information presented in Section 3 is subsequently used to identify current issues affecting the treatment of colloids in performance assessment in national programmes. These issues are identified in Section 3.3.

3.1 Finland

3.1.1 Description of concept

The KBS-3 concept for spent fuel disposal in crystalline bedrock was first introduced by the Swedish SKB in 1983 and has since been subject to research and development work both in Sweden and in Finland. At the end of the year 2012 Posiva will submit a construction license application for a repository for 9000 tonnes of spent nuclear fuel. The rock characterisation facility, ONKALO, has been constructed and will be used as the access route to the repository.

The KBS-3 concept, which is based on the multiple barrier principle, aims at long-term isolation and containment of spent fuel assemblies within durable copper-iron canisters in a way that any releases of radionuclides from the canisters are prevented for as long as they could cause any harm to people or the environment. The repository design allows for retrieval of the spent fuel, if considered necessary.

The disposal system consists of the spent nuclear fuel, the canisters, the buffer, the tunnel backfill and tunnel plugs, the geosphere and the biosphere in the vicinity of the repository. Two variants of the KBS-3 concept are being considered. The reference concept is KBS-3V (Figure 1, left), in which the disposal canisters are emplaced vertically in individual deposition holes. The alternative concept is KBS-3H (Figure 1, right), in which the canisters are emplaced horizontally in 100-300 metre long deposition drifts. Both concepts are based on one-storey underground facility with disposal tunnels at a depth of about 420 metres below ground.

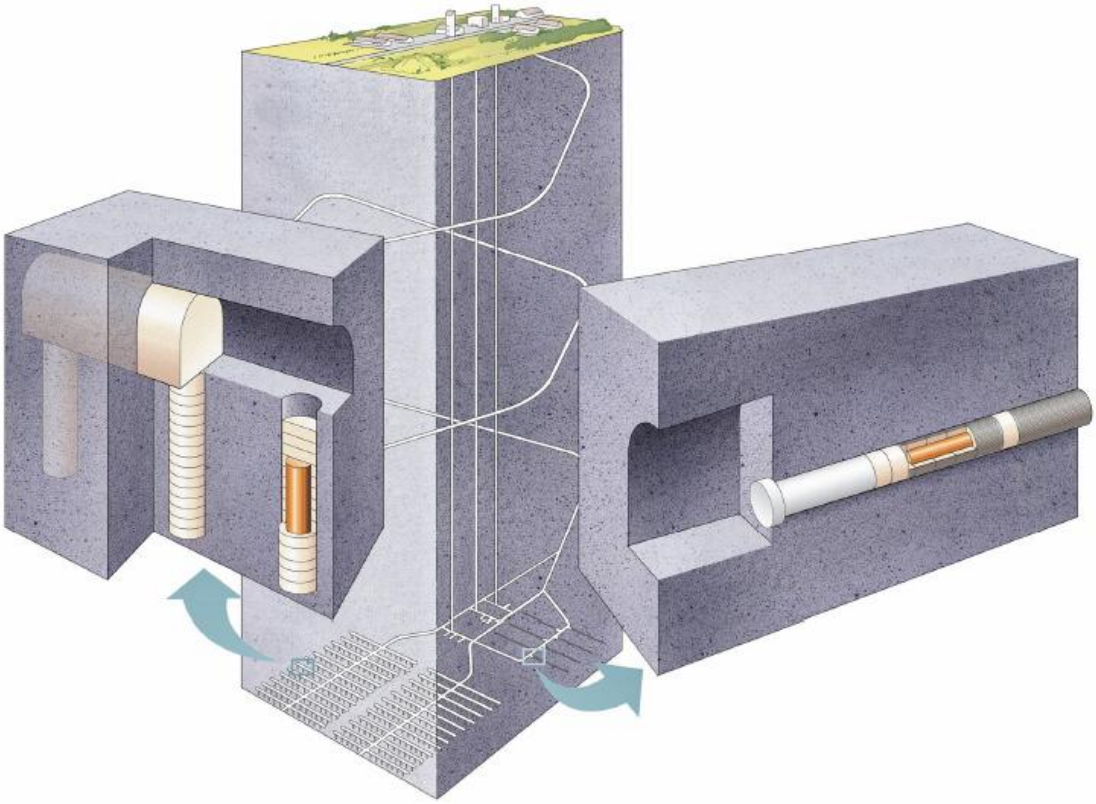

***Figure 1**. The KBS-3V (left) and KBS-3H (right) variants of the KBS-3 spent fuel disposal concept.*

3.1.2 Findings from previous studies

Posiva has carried out experimental investigations and development of a numerical erosion model to be reported in the Preliminary Safety Assessment Report portfolio in late 2012. A number of related reports are under preparation.

The investigations focussed on preparing data to validate the existing numerical model [1] in conditions geometrically similar to a constant aperture fracture intersecting a KBS-3V deposition hole.[1] The geometry is depicted in Figure 2.

[1] A more detailed description is given in section 3.2.2.

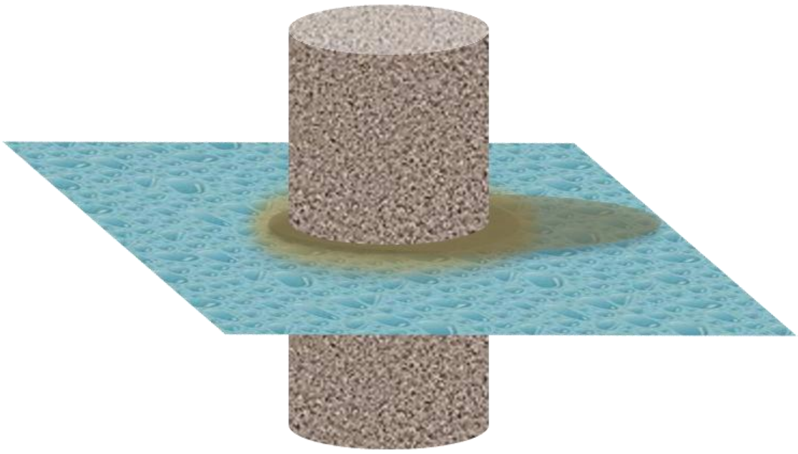

Figure 2. Depiction of chemical erosion of buffer material in an intersecting transmissive fracture. [2]

Experimental setup in 1:88-scale for artificial fracture is presented in Figure 3.

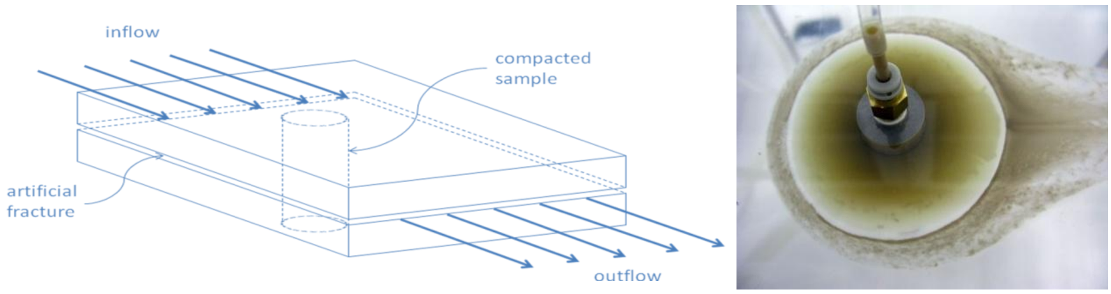

Figure 3. Schematic representation of the flow-through, artificial fracture test system (left) and a characteristic overhead image of an actual test (right). [2]

These investigations provide data to evaluate the dependence of mass loss rate on groundwater velocity, groundwater chemistry and on montmorillonite chemical composition.[2] Additionally Posiva participates in the CFM project. From this project *in situ* data on fluid dynamical properties of real variable aperture fracture have been obtained and assessed.

The findings supporting the assumptions used in SR-Site [3] evident from the currently existing data are that;

a) mass loss rate can be captured with a power-law fit so that it scales with flow velocity u as $k{\cdot}u^n$ (both k and n depend not only on geochemical conditions but on the size of the setup as well),
b) mass loss rate is directly proportional to fracture aperture ($2b_v$),
c) presence of Ca in the system results in attenuation of mass loss rate as compared to a system comprising of Na-montmorillonite and NaCl containing groundwater (attenuation of the order of 5-fold in NaCl water for 50/50 Na/Ca montmorillonite as opposed to 100/0 Na/Ca montmorillonite) and

[2] A more detailed description of these phenomena can be found within section 3.2.2.

d) mass loss ceases when groundwater salinity exceeds a stability limit observed to be between 4 to 8 mM NaCl for Na-montmorillonite.

Validation of numerical model with small scale test results is pending.

In terms of mass loss rate dependency on u^n the value for n predicted for

- buffer by experimental means is 0.27 (and k=850·b_v as in Figure 4 when mass loss rate is in kg/a),
- buffer by numerical modelling is 0.41 (and k=27.2·b_v as in Figure 5), and
- backfill by numerical modelling is 0.60 (and k=20.7·b_v as in Figure 5).

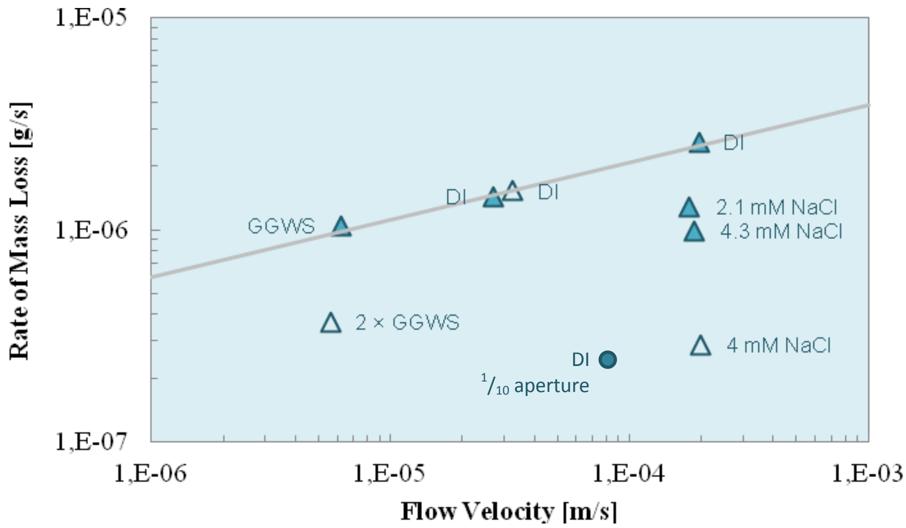

***Figure 4**. Experimentally observed mass loss rates from artificial fracture tests expressed in terms of flow velocity. Aperture was 1 mm for triangles and 0.1 mm for the circle. Filled data points are reflective of tests conducted with sodium montmorillonite and unfilled data points are reflective of tests conducted with 50/50 calcium/sodium montmorillonite. Data points are labelled with corresponding test solution composition; GGWS represents a Grimsel groundwater simulant relative to Na^+ and Ca^{2+} concentrations only (0.68 and 0.14 mM, respectively) and DI represents deionised water [4].*

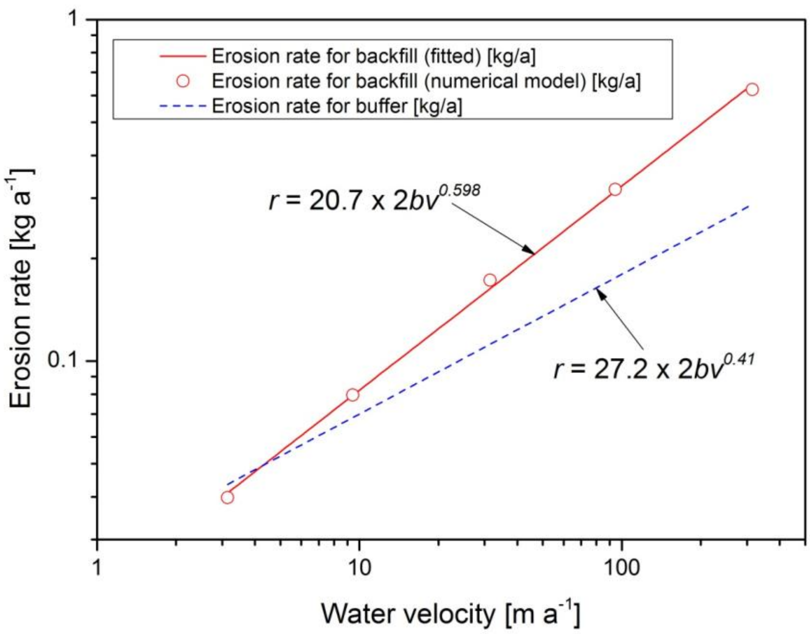

Figure 5. Montmorillonite release rates from the buffer and backfill to fractures with an aperture of 1 mm for different water velocities. The best-fit relationship for the buffer is also shown. The fit for the buffer is based on the data in Table 5-1 of Moreno et al. [5], with 2b being the fracture aperture and v being the water velocity in the fracture. [2]

These data suggest that the numerical prediction might have too strong coupling between mass loss rate and groundwater flow velocity. Moreover, experimental results (Figure 4) suggests one order of magnitude uncertainty in groundwater flow velocity having roughly the same impact on mass loss rate as the uncertainty regarding whether the salinity of groundwater is 4.3 mM NaCl in 100/0 Na/Ca montmorillonite or deionised water (DI). In this respect the same uncertainty in terms of Ca occupying half of the exchangeable cation sites in montmorillonite is clearly higher, at least few-fold and 5-fold for flow velocity v=2·10^{-4} m/s. This test series indicates that the presence of Ca has clearly greater effect on mass loss rate than the tested range of groundwater flow velocities.

As regards to the groundwater ionic strength below which mass loss occurs, the theoretically predicted limits presented in Figure 6 appear conservative when comparing to the small scale fracture tests. The theoretically predicted solution ionic strength below which a 100/0 Na/Ca montmorillonite starts to lose mass in NaCl groundwater is 25 mM as opposed to the experimentally observed 4-8 mM in the setup considered. This is still consistent with the experimental observation of stability limit of 2-4 mM ionic strength for systems in which Ca occupies 20 % of the exchanger sites as presented in Birgersson *et al.* [6].

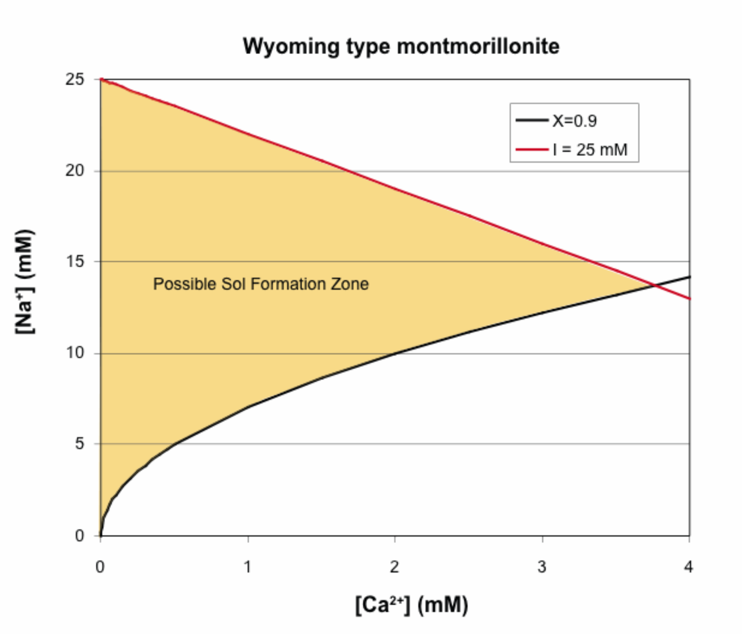

Figure 6. A possible sol formation zone for Wyoming type montmorillonite in equilibrium with external concentrations of CaCl2 and NaCl: the lower limit represents montmorillonite with a 90% calcium fraction (X) and the upper limit represents a solution ionic strength (I) of 25 mM [6].

3.1.3 Current treatment of colloids in the safety case

Colloids have received very little attention in performance assessments to date. It is felt that colloid facilitated radionuclide transport has only diminutive effects on safety. Colloids are unlikely to have any effect on the waste container integrity. Currently bentonite erosion has been omitted from the processes affecting the evolution of the near-field.

FEFTRA, ConnectFlow, REPCOM and FTRANS are all used to assess long term safety for a disposal facility (see Nykyri et al. [7] for more details). An additional tool used to represent Bentonite erosion / colloid facilitated radionuclide transport etc is Comsol Multiphysics.

Although the groundwater ionic strength and the amount of Ca in the solution does have the biggest effect on mass loss rate, the occurrence of DI conditions with no Ca in the porewater of the buffer and backfill has not been excluded. Additionally, it is expected that Na/Ca occupancy in exchanger sites in Olkiluoto repository conditions is roughly 5/3 so that the stability limit will be lower than the one measured in the small-scale tests (4-8 mM) and possibly lower than the measured one presented in Birgersson *et al.* [6] (2-4 mM). Similarly regardless of that, the effect of flow velocity appears different in the small-scale tests compared to the full-scale numerical model (the way of assessing mass loss rate more sensitive to flow velocity has been utilised since adequate validation is pending). To summarise this,

- groundwater has been assumed to be deionised, DI, at the repository level,
- all exchanger sites in clay components occupied with Na and
- mass loss rate has been assumed to scale with groundwater flow velocity as $u^{0.41}$.

Fracture aperture has been assumed to scale with the fracture transmissivity as

$$2b_v = 10 \cdot 0.0117 \cdot T^{0.33}$$

As the most expected case (termed "Reference") it has been assumed that dilute water is present during the retreat of the ice sheets, three ice sheets are formed during one glacial cycle and the ice margins are assumed to stop for 1000 years during the retreat of each of the three glaciations with melt water penetration being possible only the during summer.

The assumptions in variant 1 are otherwise the same as in the reference case except that during retreat of each of the three glaciations with melt water penetration being possible continuously.

In variant 2, dilute water is present during the growing phase of the first glaciation, which is assumed to be warm-based, with the duration of dilute conditions assumed to be equal to half of the total duration of the glaciation, and dilute water is also present as during ice-sheet retardation of the reference case.

As limits for acceptable mass loss the reasoning developed by SKB and published in SR-Site (see for example [3]) has been adopted.

The total mass loss in these kinds of conditions (as described in [2]) has been assessed to result in advective conditions in only a few deposition holes as presented in Table 1.

Case	***Inflow into deposition hole limited to <0.1 l/min***	***Inflow into deposition hole not limited***
Reference	*1*	*1*
Variant 1	*3*	*13*
Variant 2	*4*	*14*

Table 1. Number of deposition holes experiencing advective conditions due to buffer erosion during the first glacial cycle for reference and variant cases. [2]

The radiological consequences have been assessed in the case of assuming that either bentonite is poorly emplaced around an initially defective canister or that bentonite becomes ineffective with time e.g. due to the penetration of glacial melt water giving rise to mass loss. Further, it is assumed that sulphide-rich solutions will come into contact with the canister surface causing an initially small defect (1 mm) to enlarge by corrosion up to 400 mm in one step at 100 000 years after present. Thus the canister would lose its containment capacity almost entirely. The resulting total dose rate for such a scenario is presented in Figure 7 with identifier BSh-LhQ (blue curve).

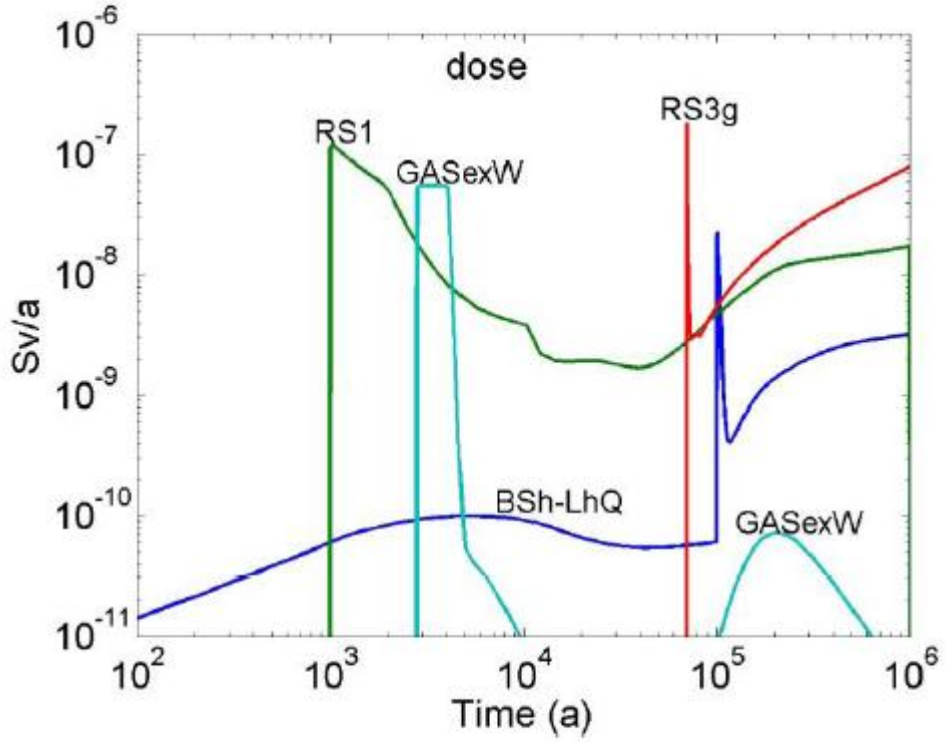

***Figure 7**. The total dose rates for selected cases of the scenarios: I) rock shear/earthquake: RS1 and RS3g, II) small hole enlarging in time due to weakened performance of bentonite: BSh–LhQ, and III) contaminated water expelled by gas from a canister: GASexW.*

Transport of radionuclides facilitated by colloids has been omitted in the latest safety analysis [7]. This is largely because as suggested by the results of the RETROCK project [8] most of the processes omitted correspond with the highest level of uncertainty and lowest level of significance associated with radionuclide transport and retardation processes in the geosphere (Figure 8). It is though recognised that in disposal systems, colloids may enhance the migration of radionuclides, provided that radionuclides become fixed to colloid particles moving at similar velocities as the groundwater does. Naturally occurring colloids can usually be neglected in performance assessments, based on their very low concentrations, but colloids formed in the repository near field may need to be considered [8].

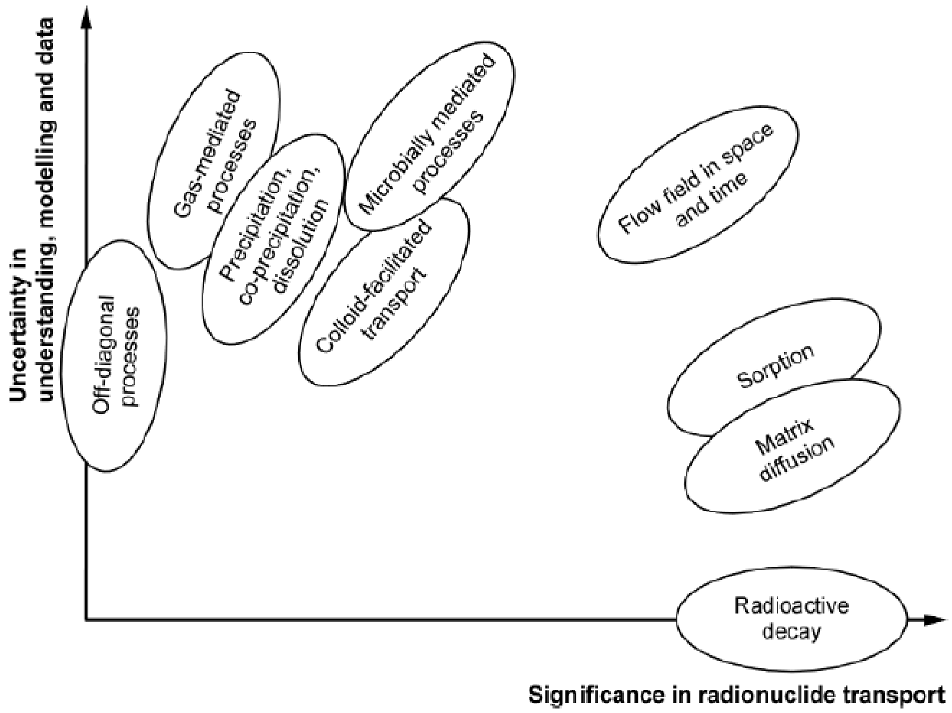

Figure 8. The uncertainties in the treatment of retention and transport processes versus their relevance for performance assessments were assessed in the RETROCK project with this kind of graph. A difficulty in placing many of these processes in a simple graph stems from their site- and concept-specificity [8].

3.1.4 Limitations and uncertainties of the current treatment

To summarise this,

- groundwater has been assumed to be deionised, DI, at the repository level,
  - since there is no data/evidences to exclude this
- all exchanger sites in clay components occupied with Na,
  - currently full scale can be assessed only by numerical modelling and the current model can only deal with monovalent systems
- mass loss rate has been assumed to scale with groundwater flow velocity as $u^{0.41}$.
  - validation of model pending so that pessimistic values are chosen
- fracture transport apertures is derived as $2b_v = 10 \cdot 0.0117 \cdot T^{0.33}$
  - justification as in Figure 9

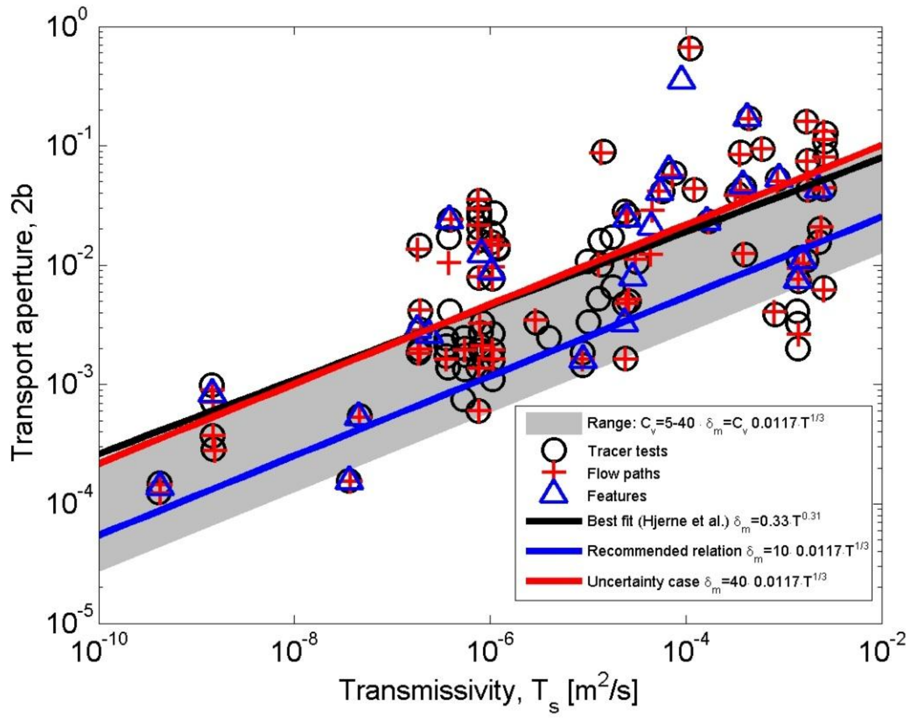

Figure 9*. *Relation between fracture transmissivity and volume aperture. Based on digitised data from Hjerne et al. [9, Figure 6-1].

- Formation of accessory mineral bed layers during erosion of bentonite is neglected
  - there are indications of such a process but only few qualitative evidences
- erosion will be significant only in a few locations
  - result of assessment as outlined above
- radiological consequences are small
  - see Nykyri *et al.* [7]

3.2 Sweden

3.2.1 Description of concept

Several decades of research and development has led SKB to put forward the KBS-3 method for the final stage of spent nuclear fuel management. In this method, copper canisters with a cast iron insert containing spent nuclear fuel are surrounded by bentonite clay and deposited at approximately 500 metres depth in saturated, granitic rock, see Figure 10. The purpose of the KBS-3 repository is to isolate the radioactive waste from man and the environment for very long times. Around 12,000 tonnes of spent nuclear fuel is forecasted to arise from the currently approved Swedish nuclear power programme (the last of the 10 operating reactors is planned to end operation in 2045), corresponding to roughly 6,000 canisters in a KBS-3 repository.

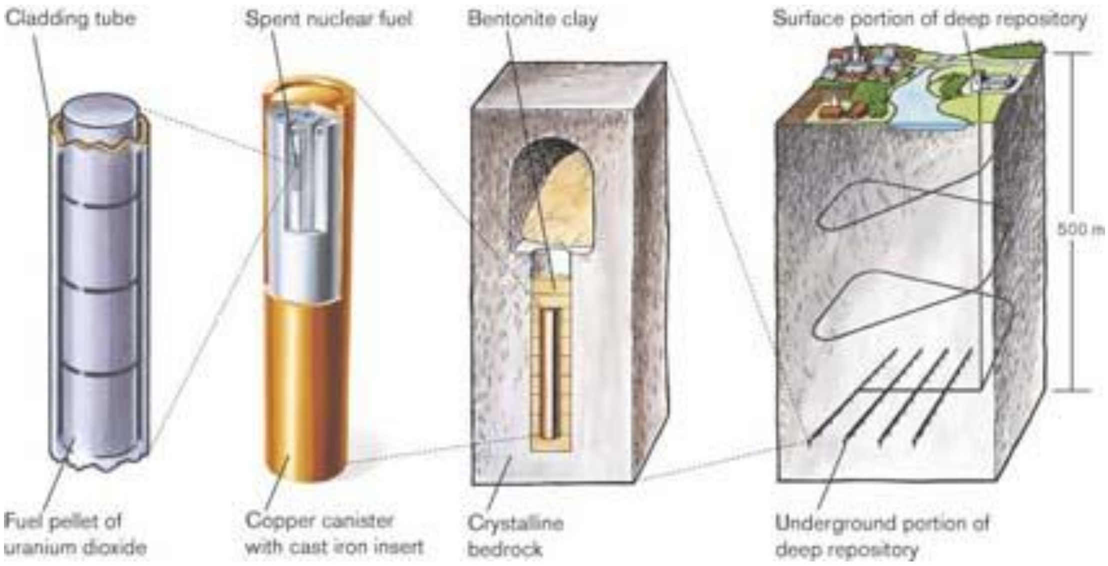

Figure 10. The KBS-3 concept for disposal of spent nuclear fuel.

3.2.2 Findings from previous studies

SKB has carried out an internal project (Bentonite Erosion) to study erosion of bentonite in dilute waters (“dilute” meaning representative of glacial meltwaters). The purpose of the project was to construct a quantitative model for judging the extent of the erosion process in the SR-Site safety assessment. The project ran from 2007 to 2009. The different phases of the project were literature studies, modelling and experiments. There were three phases of this work as listed below:

- Literature studies and information gathering [10]
- Modelling [1, 6 and 11]
- Experiments [6, 12 and 13]

3.2.3 Current treatment of colloids in the safety case

3.2.3.1 Bentonite Erosion

Erosion in KBS-3 for a single canister position was treated in SR-Site. The key factors determining whether erosion will have an effect on the performance of the repository are:

1) The conditions when colloid release can occur
2) The mass loss from erosion
3) The allowed mass loss before the diffusion barrier is lost

The handling of these factors in SR-Site [3] is briefly summarised below.

3.2.3.2 Conditions when colloid release can occur

Based on the experimental studies of Birgersson et al. [6], water with cation content higher than 2–4 mM charge equivalents is considered able to prevent colloidal sol formation provided that the calcium content in the montmorillonite is above 20%, irrespective of montmorillonite type. In SR-Site, the upper limit of 4 mM was selected as a pessimistic limit. This criterion is also in agreement with reported amounts of calcium salts needed to coagulate initially homo-ionic Na-montmorillonites [14, 15, and 16]. In equilibrium with typical Forsmark water the bentonite is expected to have an approximately equal population of calcium and sodium ions in the exchanger. Ion exchange processes during the evolution of the repository may alter the counterion content relative to the initial state.

3.2.3.3 Quantification of buffer loss

Figure 11 shows a fracture intersecting the canister deposition hole which is filled with compacted bentonite. The bentonite, when wetted, swells out into the fracture. It has a very high swelling pressure when highly compacted but the swelling pressure decreases with decreasing bentonite density.

The smectite particles are pulled and pushed into the water that seeps in the fracture by the different forces acting on the particles. If the porewater cation concentration is below 4 mM charge equivalents, the particles at the bentonite/water interface can swell/diffuse into the moving water and be carried away. There is also a region where the gel/sol has so low a particle concentration that it is little more viscous than water and can flow away.

The loss of particles is thus influenced both by particle diffusion and by advective flow of the dilute gel/sol. For both mechanisms, the flow rate of water and gel in the fracture will set the total rate of loss.

A DLVO (Derjaguin and Landau, Verwey and Overbeek) based force-balance model for spherical colloids [17] has been adapted to parallel clay layers [1, 10] and used to calculate the swelling of Na-montmorillonite into fractures filled with water of low ionic strength. The force-balance model uses DLVO to describe swelling pressure and a Kozeny-Karman-like expression fitted to experimental results to describe hydraulic conductivity. The model is adjusted and tested against free-swelling experiments with results obtained through magnetic resonance imaging (MRI) [12]. Advective loss of montmorillonite is modelled by combining the force-balance model for swelling with a viscosity model for the repulsive montmorillonite gel and the Darcy equation for two-dimensional flow in a fracture intersecting the deposition hole.

The transient expansion is not considered. The montmorillonite release rate, $R_{Erosion}$, is found to be proportional to the water velocity, v, to the power 0.41 and directly proportional to the aperture, δ, according to:

$$R_{Erosion} = A \cdot \delta \cdot v^{0.41}$$

where A = 27.2 is a constant yielding the loss rate in kg/yr when the water velocity is given in m/yr and the aperture in m.

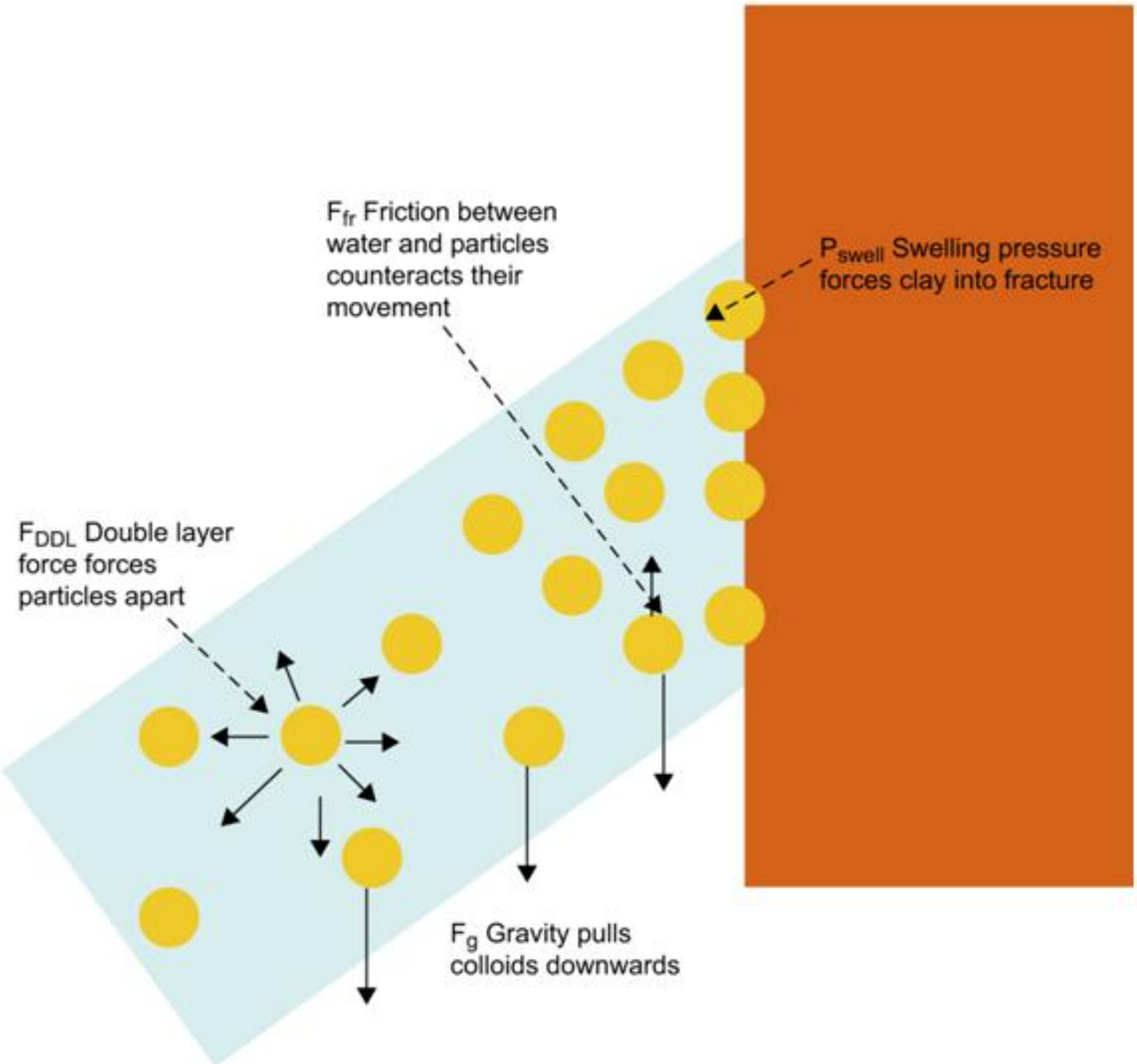

Figure 11 Schematic of forces acting on the bentonite in a deposition hole and fracture (diffuse double layer, Van der Waals (not shown), friction in water and forces of gravity) after [1]. Friction forces against fracture surface are not shown

The model assumes that the bentonite consists of only montmorillonite which is converted to a pure Na-form and the buffer porewater is assumed to be depleted of Ca^{2+} ions in the fracture buffer interface. This means that Ca^{2+} ions for the prevention of colloid sol formation will not be supplied by the buffer.

3.2.3.3.1 Allowed mass loss before the diffusion barrier is lost

The swelling and sealing of bentonite cannot take place unhindered since there is a resistance to swelling caused by friction both internally in the bentonite and between the bentonite and the surrounding fixed walls represented by the rock surface and in some cases the canister. In order to investigate how well the buffer material seals the openings resulting from the mentioned processes a number of finite element calculations with the code Abaqus have been performed [18].

The case that has been modelled represents a huge loss of bentonite after a long time of erosion where one to three bentonite rings at the upper end of the canister are missing.

The rings are 50 cm thick and are for the sake of the calculations assumed to be missing from the installation. This case represents an extreme loss of bentonite by colloid erosion. The calculations comprise cases with an empty space of 0.5 m, 1.0 m and 1.5 m. Figure 12 shows the course of swelling for the base case (two missing rings) assuming a friction angle Φ = 8.69°. After a rather long time the space is almost completely filled with bentonite but there is a small remaining final opening and the void ratio is rather high (1.7) close to that opening. The density is so low that the expected swelling pressure might very well be below 100 kPa at the position closest to the canister.

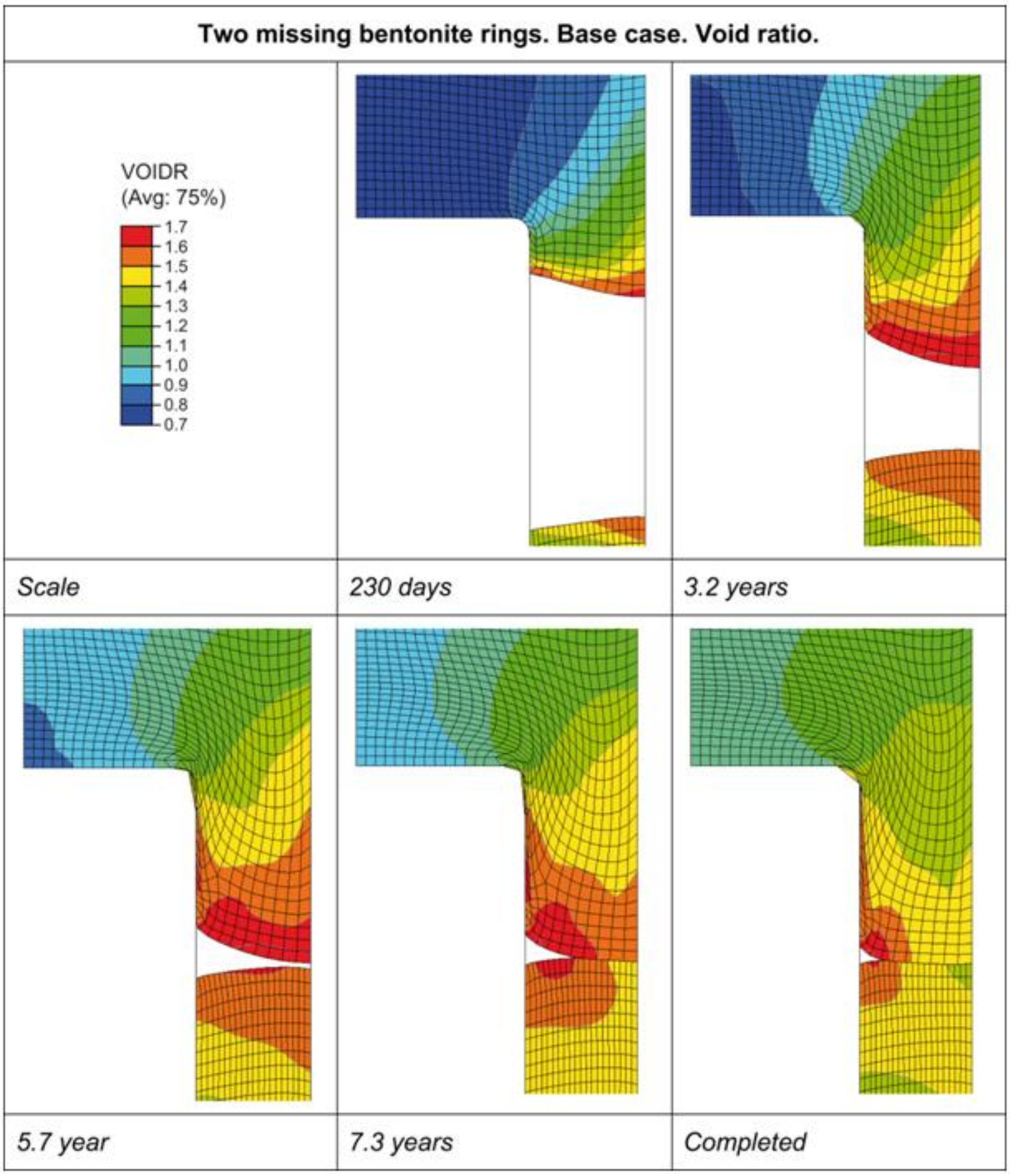

Figure 12 Void ratio plotted at different times for the base case with two missing rings [18]

Advective conditions in the buffer can occur if the hydraulic conductivity is sufficiently high. The buffer function indicators prescribe a hydraulic conductivity of 10^{-12} m/s and a swelling pressure of 1 MPa to rule out advection in the buffer. These values do, however, have significant safety margins included in them. To ensure that the self-sealing ability is maintained and no channels or pipes will be formed, a certain swelling pressure is required. The minimum swelling pressure needed will be about 100 kPa. This is based on laboratory investigations in which piping has been observed at ~60 kPa [19]. To ensure this for all expected groundwater compositions, a minimum dry density of 1,000 kg/m3 is required. This corresponds to a void ratio of 1.75. As seen in Figure 12 this requirement is still met in almost the entire buffer diameter when two entire bentonite rings are omitted, corresponding to a dry mass loss of 2,400 kg. For the case when the buffer erodes by colloid formation, the mass loss may be more local compared with the case in which entire blocks are omitted and it is more appropriate to consider the corresponding limit for losses over typically half the circumference, i.e. 1,200 kg. At higher mass losses, the swelling pressure cannot be guaranteed and advection in the buffer has to be considered.

3.2.3.4 Colloid-facilitated transport

The presence of bentonite material in tunnel backfill and borehole buffer is expected to result in bentonite colloids in the groundwater near deposition holes and along the geosphere transport pathways. Radionuclides that have a strong affinity for bentonite will sorb onto bentonite colloids and may be transported through the geosphere with reduced interaction with the rock matrix, i.e., with a reduced retention. Colloid facilitated transport involves a complicated combination of processes, many of which can mitigate the transport. Mitigating processes include colloid retardation in fractures, physical filtration (straining) of colloids in fractures, colloid flocculation and sedimentation, saturation of sorption sites on colloids, and competition for sites on colloids. These processes are uncertain or involve uncertain parameters that are difficult to quantify in short duration experiments. Rather than attempting to develop detailed process models for colloid-facilitated transport, potential mitigating processes are ignored so as to place an upper bound on the possible effect. Ignoring these potential mitigating processes and taking into consideration that sorption of radionuclides onto bentonite is understood to be a reversible process on the time scale of geosphere transport, the effect of colloids in facilitating transport may be modelled through the introduction of effective transport parameters [20].

Using the MARFA transport code and the effective parameters, the effects of colloids are demonstrated in Figure 13. Except for the effects of colloids, this modelling case is identical to the central corrosion base case in SR-Site. The dark blue curve is near-field mean annual release expressed as an effective dose; the black curve is the far-field mean-annual effective dose without colloids. The green and pink curves are the far-field doses with colloid concentrations of 10 mg/l and 10 g/l, respectively.

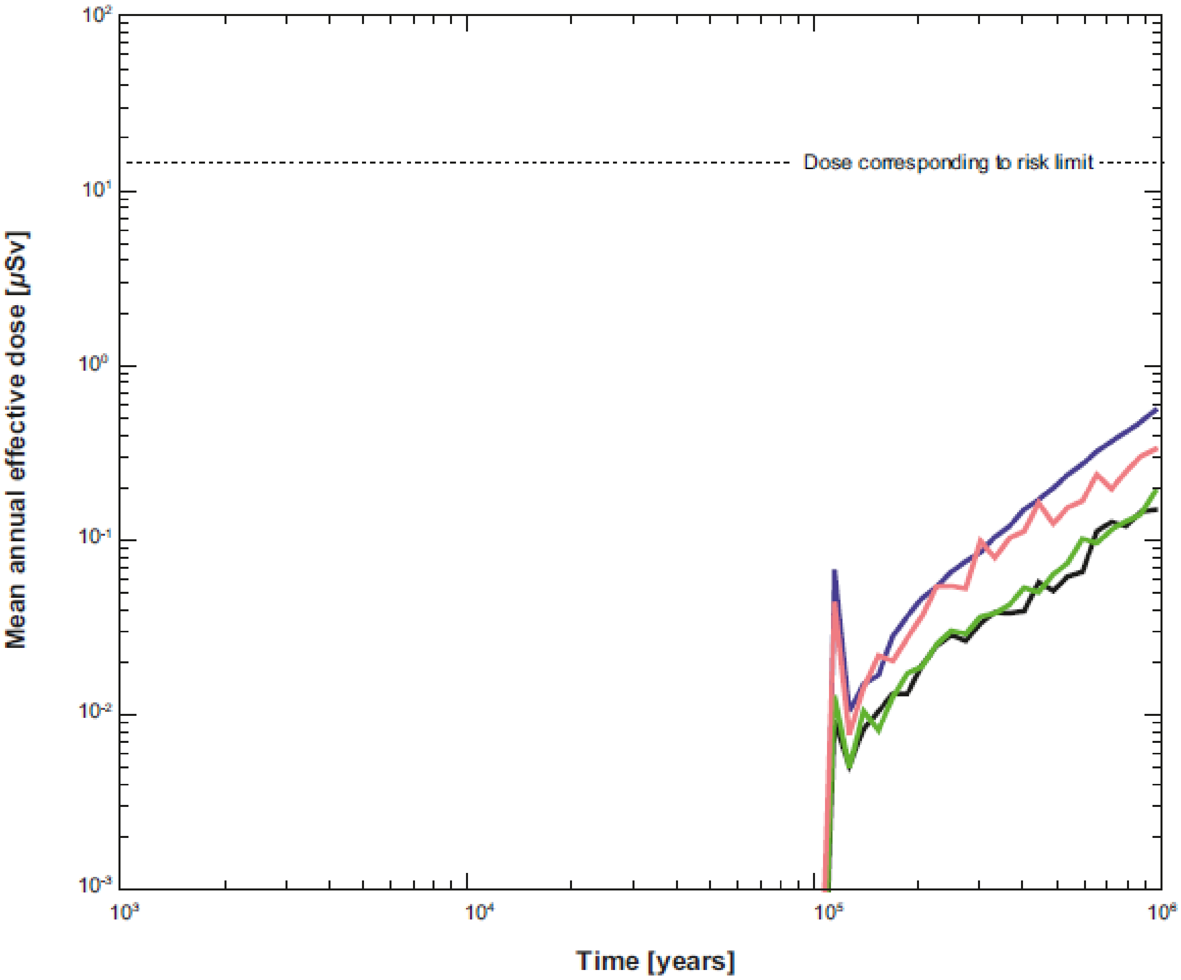

Figure 13. Mean annual effective dose for the probabilistic central corrosion case, with and without the effects of bentonite colloids in facilitating transport. The black, green, and pink curves are far-field dose with colloid concentrations of 0 mg/l, 10 mg/l, and 10 g/l, respectively. The dark blue curve is the near-field dose equivalent release [20].

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

When the buffer is partially or completely eroded, a cavity filled with a slurry of water containing colloidal clay particles may exist in the deposition hole. Should the canister be breached under such circumstances, the clay particles are not expected to affect the fuel dissolution rate. Dissolved U(IV) would, however, be expected to sorb strongly to the clay particles. This sorption increases the amount of U(IV) released in solution from the re-precipitated UO_2(s) or the fuel matrix. In this case, the amount of U(IV) sorbed on clay particles may be calculated as the *Kd* value for U(IV) on clay particles multiplied by the U(IV) concentration in solution, determined by UO_2(s) solubility.

No limit to the U(IV) release rate from UO_2(s) to satisfy U(IV) solubility limits in the canister void is then posed. In the case when all re-precipitated UO_2(s) is dissolved due to sorption to clay particles the remaining U(IV) needed to saturate clay particles is released from the fuel matrix, resulting in an increase of the fuel dissolution rate. Fuel dissolution in the presence of a clay slurry needs to be considered.

The eroded void volume of the deposition hole is assumed to be filled with clay particles that enhance the dissolution rate of the UO_2 fuel matrix. The outward transport rate of U, R_U (mole/yr), is then obtained as:

$R_U = C_{Sol}U \cdot q \cdot (1+C_{Clay} \cdot K_d)$
where
$C_{Sol}U$ is the solubility of U(IV) (mole/m^3),
q is the advective flow at the deposition hole (m^3/yr),
C_{Clay} is the concentration of clay in the flowing fluid (kg/m^3),
K_d is the partitioning coefficient between solid phase and solution for U in a clay slurry (m^3/kg).
Using C_{Clay} = 10 kg/m^3 and distributions of K_d, $C_{Sol}U$ and q for the central corrosion case in SR-Site [3], the contribution from U transport on clay particles to the fuel dissolution rate is calculated probabilistically and compared to the probabilistically sampled ordinary fuel dissolution rate for the central corrosion case, see Figure 14.

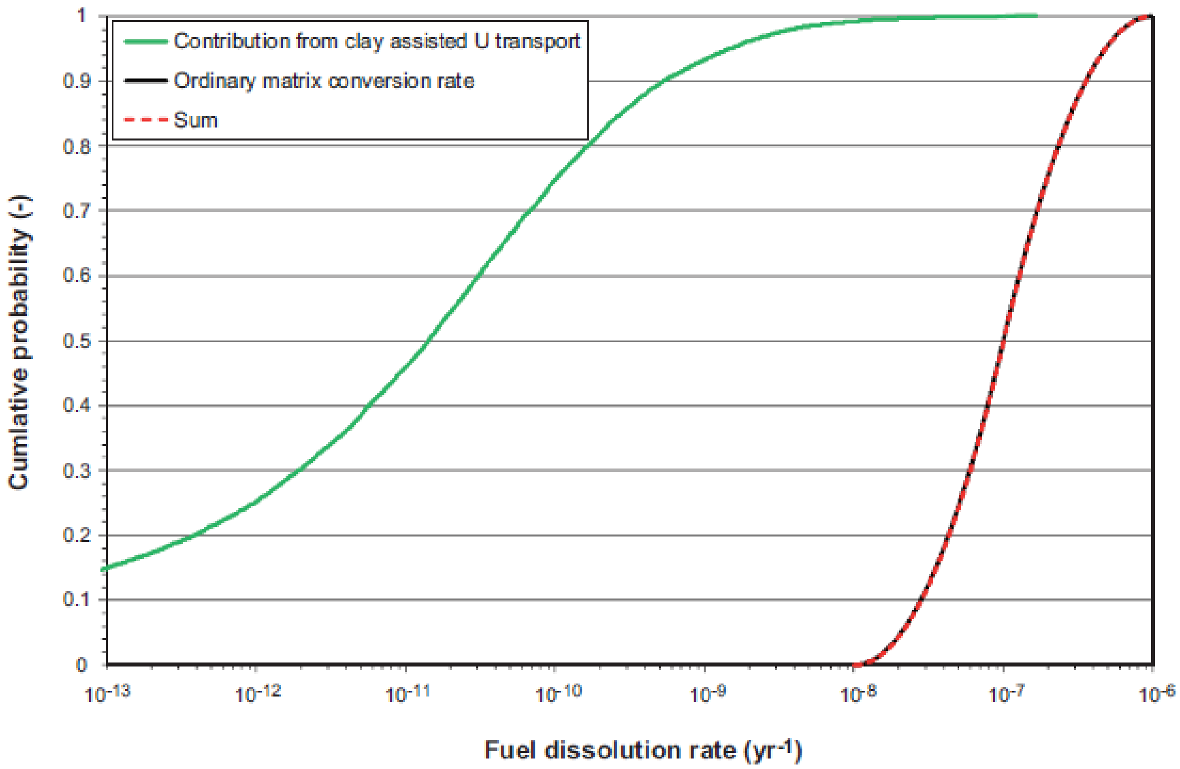

Figure 14 Cumulative distributions of fuel dissolution rate. The contribution from clay assisted U transport has a negligible impact on the distribution for the ordinary matrix conversion rate [3]

The following is noted.

- This process is only active during the relatively limited periods when the deposition position is exposed to dilute groundwater such that clay colloids can form.
- Inflowing groundwater may have U concentrations exceeding 10^{-9} mol/L, but the influence of the occupation of sorption sites of clay particles by natural uranium present in the groundwater is pessimistically neglected.
- Should sorption to clay colloids be modelled as irreversible and assuming that all sorption sites were to be occupied by U, then a transport rate of the order of $2.5 \cdot 10^{-2}$

moles/yr is calculated. Irreversible sorption, as well as U occupancy of all available sites is, however, ruled out for the conditions in the deposition hole.

3.2.4 Limitations and uncertainties of the current treatment

In SR-Site, for situations where the groundwater has a total positive charge of less than 4 mM, the loss of bentonite is calculated with the model developed [1]. The model can be applied both to the buffer and to the backfill. However, there are a number of uncertainties associated with this treatment.

- The knowledge concerning colloid sol formation and colloid stability is good concerning the effects of mono- and divalent ions. However, modelling of the correlation effects caused by divalent ions is demanding. Since the model basically is based on monovalent ions the calculated loss should be pessimistic.
- The model does not consider face to edge interactions between bentonite platelets. Not considering these interactions probably leads to an overestimation of the loss.
- Filtering effects by accessory minerals could potentially limit or even eliminate the release of colloids from the buffer. However, this is disregarded in the current treatment of the process, since there is a lack of evidence that efficient filters actually do form.
- In the model, the expansion has been taken to be horizontal, thus neglecting gravity. Scoping calculations [1] suggest that gravity will give small effects as, in the model, the smectite sheets have separated into essentially individual colloid particles. For gravity to have an effect the particles must be considerably larger.
- The concentration limit for cation charge is only based on experimental observations.

Most of the uncertainties are treated with pessimistic assumptions, leading to the conclusion that advective transport conditions in the buffer do not need to be considered in any of the deposition holes during the initial temperate period.

3.3 United Kingdom

3.3.1 Description of concept

3.3.1.1 UK Managing Radioactive Waste Safety Programme

In June 2008, the UK Government published the Managing Radioactive Waste Safely (MRWS) White Paper, entitled "*A Framework for Implementing Geological Disposal*" [21] (hereafter referred to as the White Paper) – this established the implementation programme for a UK Geological Disposal Facility[3] (GDF).

The White Paper defines the UK Government's framework for managing higher-activity radioactive waste in the long-term through geological disposal and the role of the RWMD as the effective delivery organisation of the NDA for the implementation of geological disposal.

The White Paper also sets out the treatment for the following strategic uncertainties:

- The design and layout of the GDF, both above and below ground will be tailored to the Baseline Inventory, defined in the White Paper, and the characteristics of the site in question.

- The UK Government sees no case for having more than one GDF if a single facility can be developed to provide suitable, safe containment for the Baseline Inventory.

[3] Although referred to here, and elsewhere, in the singular, the term "a geological disposal facility" does not mean that the decision to build a single facility has been taken. UK Government has expressed a preference for a single co-located geological disposal facility but multiple facilities have not been ruled out.

- Planning, design and construction of the GDF should be carried out in such a way that the option for extended retrievability is not excluded[4].

However, significant uncertainties still exist and need to be recognised and understood, these uncertainties include:

- The inventory of radioactive wastes and materials (e.g., radionuclide content, physical form and condition, and volumetric uncertainties);
- The waste package design for each waste type;
- The GDF location and geological environment (determined by the volunteer community);
- The disposal concept.

3.3.1.2 Inventory

The White Paper defines the Baseline Inventory as an estimate of the higher-activity radioactive waste and other materials that could, possibly, come to be regarded as wastes that might need to be managed in the future through geological disposal. It does not include

Low Level Waste (LLW) suitable for disposal at the existing Low Level Waste Repository (LLWR) facility in Cumbria, but does include materials not at present classified as waste such as spent nuclear fuel or uranium and plutonium stocks.

The Baseline Inventory is based on UK-wide inventory data so includes wastes that are expected to be managed under the Scottish Government's policy of interim near-surface storage and not through geological disposal. The Baseline Inventory was summarised in the White Paper and is provided in Table 2.

Table 2: 2007 Radioactive Waste and Materials Inventory [21]

Materials	**Packaged volume**		**Radioactivity** (At 1 April 2040)	
	Cubic Metres	%	**Terabequerels**	%
HLW	1,400	0.3%	36,000,000	41.3%
ILW	364,000	76.3%	2,200,000	2.5%
LLW (not for LLWR)	17,000	3.6%	<100	0.0%
Spent nuclear fuel	11,200	2.3%	45,000,000	51.6%
Plutonium	3,300	0.7%	4,000,000	4.6%
Uranium	80,000	16.8%	3,000	0.0%
Total	**476,900**	**100**	**87,200,000**	**100**

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3.3.1.3 GDF Location and Geological Environment

The range of geological environments that could be suitable for hosting a geological disposal facility for higher-activity radioactive wastes in the UK is diverse. Government has committed to undertaking a volunteer approach to siting a geological disposal facility, which implies that the geological environment available for the disposal facility will depend on the location of the volunteer communities. The approach that NDA RWMD will take until such time as more specific information becomes available is to define a limited number of generic geological

[4] The Government's view is that the decision about whether or not to keep a GDF (or vaults within it) open for an extended period can be made at a later date in consultation with the independent regulators and local communities.

environments, encompassing typical UK geologies, for use in engineering designs and in the safety and environmental assessments that underpin the Disposal System Safety Case[5] (DSSC) for a UK GDF.

The defined generic geological environments, developed within NDA RWMD, are based on consideration of the host rock formation (where waste emplacement will take place) and the cover rocks (the geological formations that occur between the host rock formation and the ground surface). The generic geological environments are not associated with any specific parameters or characteristics other than those that could be implied from the geological characteristics of the formations. The descriptors and groupings for these rock types are necessarily at a high level to reduce the number of options to a manageable number.

3.3.1.4 Geological Disposal Concepts

A range of generic geological disposal concepts is available that can provide safe and secure geological disposal options for any suitable UK geological environment, although it will be necessary to consider different disposal concepts for different geological environments.

Work carried out by NDA RWMD in 2008 [22, 23] reviewed the range of possible concepts for geological disposal of ILW, HLW and SF in the UK. The work drew on previous work in the UK, and disposal programmes in other countries, to identify disposal options for generic geological environments (rock formations and their surrounding geological setting).

For the engineering designs and the safety and environmental assessments that underpin the DSSC, the approach that will be adopted to managing this uncertainty in the disposal concept will be to identify a limited number of options, that is, a “catalogue of concepts” approach. To use available resources effectively, illustrative concepts will be developed for each of the three generic geological environments. However, this does not mean that any of the illustrative concepts developed will necessarily be the concept used in that geological environment – at this stage, no geological disposal concepts have been ruled out.

The illustrative geological disposal concept examples have been selected by:

- Consideration of the concepts identified in the Geological Disposal Options studies for ILW/LLW and HLW/SF [5, 6].
- Basing the illustrative examples on geological disposal concepts that have previously been developed in the UK or are being developed in national programmes in other countries.
- Using the criteria developed previously to select KBS-3V as an example reference HLW/SF Concept, that is, the concept must be well-developed and supported by extensive R&D and have been subject to detailed safety assessment; regulatory scrutiny and international review (see Reference [24]).

The illustrative concept examples selected are listed in Table 3 and the attached notes present the key reasons why these examples were selected in preference to other possible disposal concepts. These illustrative concept examples will be considered at a high level in the DSSC and information from them will be placed in a UK context (e.g., concept designs will be scaled for the UK Baseline Inventory and key concept issues will be interpreted for UK specific factors).

The post-closure safety assessments will also take into consideration the impact of different flow characteristics in the host rock and the variability in the overlying geological formation.

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[5] The DSSC is an integrated safety case encompassing transport of waste to a disposal facility, construction, operation and long-term safety case for UK higher-activity wastes unsuitable for near surface disposal. The DSSC will also be developed in stages during the regulatory approval process affecting the GDF, which will span many years. The DSSC, and the supporting safety reports, also form a focus for continued dialogue and consultation with the regulators, public and other stakeholders throughout the project.

Table 3: Illustrative Disposal Concept Examples

Host rock	Illustrative Geological Disposal Concept Examples[d]	
	ILW/LLW	HLW/SF
Higher strength rocks[a]	UK ILW/LLW Concept *(NDA, UK)*	KBS-3V Concept *(SKB, Sweden)*
Lower strength sedimentary rock[b]	Opalinus Clay Concept *(Nagra, Switzerland)*	Opalinus Clay Concept *(Nagra, Switzerland)*
Evaporites[c]	WIPP Bedded Salt Concept *(US-DOE, USA)*	Gorleben Salt Dome Concept *(DBE-Technology, Germany)*

Notes

a. Higher strength rocks – the UK ILW/LLW concept and KBS-3V concept for spent fuel were selected due to availability of information on these concepts for the UK context.

b. Lower strength sedimentary rocks – the Opalinus Clay concept for disposal of long-lived ILW, HLW and spent fuel was selected because a recent OECD Nuclear Energy Agency review regarded the Nagra (Switzerland) assessment of the concept as state of the art with respect to the level of knowledge available. However, it should be noted that there is similarly extensive information available for a concept that has been developed for implementation in Callovo-Oxfordian Clay by Andra (France), and which has also been accorded strong endorsement from international peer review. Although we will use the Opalinus Clay concept as the basis of the illustrative example, we will also draw on information from the Andra programme. In addition, we will draw on information from the Belgian super container concept, based on disposal of HLW and spent fuel in Boom Clay.

c. Evaporites – the concept for the disposal of transuranic wastes (TRU) (long-lived ILW) in a bedded salt host rock at the Waste Isolation Pilot Plant (WIPP) in New Mexico was selected because of the wealth of information available from this United States Environmental Protection Agency (EPA) certified, and operating facility. The concept for disposal of HLW and spent fuel in a salt dome host rock developed by DBE Technology (Germany) was selected due to the level of concept information available.

d. For planning purposes the illustrative concept for depleted, natural and low enriched uranium is assumed to be same as for ILW/LLW and for plutonium and highly enriched uranium is assumed to the same as for HLW/SF.

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These high-level examples will be investigated in more detail than was covered in the 2008 Options Studies. The illustrative concepts will be used to:

- Support the scoping of the impacts of a GDF.
- Develop the disposal system specification, engineering design and safety case methodology.
- Support prioritisation of the R&D programme.

The example for strong rocks will be assessed in more detail in order to support the Letter of Compliance process[6]. This example was selected for more detailed analysis due to the availability of information for the UK context (although the HLW/SF concept was not originally UK based, work has previously been carried out to provide information appropriate to the UK context [24]).

Given the on-going MRWS programme, and the role that NDA RWMD fulfils in this programme, the safety case-related studies NDA RWMD is undertaking are geology, site and disposal concept generic.

3.3.2 Findings from previous studies

Colloids are a site-specific issue which will need to be studied at the appropriate level of detail once a UK site or sites have been identified. In the meantime NDA RWMD draws on previous work conducted by the former Nirex and work conducted by other international programmes.

[6] The UK Letter of Compliance process concerns the disposability assessment of proposed waste packages in order to determine their suitability for eventual geological disposal.

Nirex previously had a programme of work to investigate a potential site in the Sellafield area of Cumbria. A set of calculations of the potential impact of colloid transport on repository performance was conducted as part of this work and is summarised in [25]. The report describes the results of some deterministic calculations to address uncertainties concerning the behaviour of colloidal particles and their potential impact on radiological risk arising from a deep repository for radioactive waste. The calculations are based on the assessment models developed for Nirex 97, a post-closure performance assessment of a deep repository at Sellafield, in which the presence of colloids was treated as a key bias.

As a result of the HLW/SF concept being a relatively recent addition to the UK GDF programme, historic work on colloids has largely been focussed on ILW/LLW repository concepts where colloids would be generated largely from cement in the near field of the GDF (see for example [26, 27, 28, 29, 30]). Although there will need to be some bentonite specific research onto colloids, there will be some areas where our studies onto cementitious colloids will be transferable and applicable to generic colloid issues.

The potential erosion of bentonite colloids from the backfill and buffer has already been highlighted as an example where colloid mobilisation may be important. This process has been under examination in the CRR (colloid and radionuclide retardation) and CFM (colloid formation and migration) experiments in the Grimsel Test Site in Switzerland (see [31,32,33] for details) and the Colloid Project in the Äspö URL in Sweden [34].

For disposal concepts that incorporate a bentonite buffer surrounding the waste packages, colloids are not expected to be mobile out of the near field because an intact buffer will provide an effective barrier to colloid migration [35]. However, there remains significant uncertainty concerning the potential for colloid generation from the buffer by erosion; severe erosion, or other chemical changes to the buffer and its associated porosity [36], could lead to loss of barrier function. For such potential scenarios, the impact of near-field colloids needs be considered in more detail in PA.

3.3.3 Current treatment of colloids in the safety case

The DSSC considers both quantitative and qualitative inputs to the safety case: qualitative case arguments, which serve to demonstrate a depth of understanding of (in this case) the issue of GDF-derived colloids and their potential consequences, will be important in the safety case as the output of quantitative modelling. Such safety case arguments could be derived from site specific understanding and knowledge, and also from analogue studies that draw on information both from other sites that are being considered internationally for a GDF, and from other scientific and engineering disciplines.

They were several high level assumptions made as part of the Assessment of the Post-closure Performance of a Deep Waste Repository at Sellafield [37]. Colloids were assumed to be stable and to travel at the same velocity as the groundwater. Radionuclides were assumed to sorb reversibly onto the surfaces of the colloids in both the near and far fields with linear distribution coefficients. These assumptions will be revisited in future post-closure safety assessments (PCSA) taking into account international best practice in the area.

Diffusion through the bentonite buffer for the HLW illustrative geological disposal concept example (see Table 3) was not a parameter varied as part of recent PCSA calculations [38] although this may be a variable in future calculations.

Methodologies for treating colloids in performance assessment (PA) were developed [25, 37] in support of the Nirex 97 assessment of a deep repository at Sellafield, in which the potential impact of colloids was considered as a potential bias. The appropriateness of the approach used would need re-assessment in future work to which BELBaR is contributing.

There is uncertainty concerning the reversibility of sorption processes to colloids in general (as mentioned this was an assumption in [37]). The treatment of near-field colloids developed for the Nirex 97 assessment, in which the near field is treated as a homogeneous well-mixed system with a uniform distribution of stable and mobile colloids in the transport porosity, requires no assumptions to be made concerning the reversibility of radionuclide uptake to colloids [39].

One calculation was performed in [25] using highly pessimistic assumptions to illustrate the possible impact of irreversible sorption of radionuclides to colloids in the far field. The peak risk arising from 36Cl and 129I after 40,000 years is not sensitive to the presence of near- or far-field colloids because chlorine and iodine are not solubility limited in the near field and are weakly sorbing and iodine are not solubility limited in the near field and are very weakly sorbing. Colloids may impact upon the risk arising from strongly sorbed radionuclides such as 99Tc and the actinides [26].

Thus the approach provides a suitably cautious method for scoping the impact of colloids in the near field on the source term that includes the irreversible sorption case to mobile colloids. Only if such scoping calculations indicate that the impact of the near-field colloid population is potentially significant would consideration of colloids in more complex near-field models be justified. [7]

3.3.4 Limitations and uncertainties of the current treatment

Detailed studies (colloid-rock interactions) are considered more appropriate at a concept and site(s)-specific level. In the absence of any site(s) for consideration as a potential for a GDF for UK higher activity wastes, the studies undertaken by NDA RWMD are generic and consider a range of UK-relevant geologies and UK-relevant disposal concepts. Use is therefore made of generic geological environments as a basis for studies (see e.g. [38, 40, 41] and references therein); existing site-specific data (national and international, including data relevant to specific underground research laboratories) can be used additionally as a learning aid - the knowledge gained from the use of generic and example datasets is anticipated to be useful in later, site-specific, stages of the MRWS programme.

The assumption of sorption reversibility is an important issue. In some situations to assume reversibility of sorption (onto the rock or near-field materials) is pessimistic because the radionuclide concentration dissolved in the groundwater and the amount of radionuclide that may be sorbed onto colloids may be overestimated. On the other hand, irreversibility of radionuclide sorption onto colloids could increase radionuclide mass transport. The understanding of the reversibility of sorption of radionuclides to colloid surfaces needs to be improved to allow better representation in the safety case (i.e. rates of adsorption/desorption processes). This is likely to be conducted once a UK site(s) has been identified.

If sorption to colloids is found to show significant irreversibility and colloid particles are found to persist on a site specific basis, then there may be a need to develop further understanding of colloid mobility and chemical stability in the near field and into the far field [39]. However, the same processes by which radionuclides may be irreversibly sorbed to colloids are also likely to occur at the surfaces of cements and rock. A key conclusion of [39] stated that in reality given that near-field colloids are expected to have limited persistence (i.e. finite lifetimes) in the geosphere, the effects of radionuclide retardation by immobilisation with solid phases are likely to outweigh the detrimental effects of transport of radionuclide irreversibly bound to near-field colloids. This is an area which will need to be investigated and understood during the site-specific stage.

[7] Note: The risk arising from some calculations in [25] are over the risk guidance level of 10^{-6}/year and RWMD would therefore need to re-assess whether or not the models conservatisms were appropriate.

4 Synthesis of issues

Information presented in Section 3 was used to identify current issues affecting the treatment of colloids in performance assessment in national programmes. These issues are identified in Section 4 in a series of tables.

Note that the relevance of the information contained in these tables to national programmes varies on a programme by programme basis.

The issues are linked in these tables as appropriate to the Work Packages considered in the BELBaR project (see Section 2 for further details), clarifying how the issues raised by national programmes regarding the treatment of colloids in performance assessment will be progressed by the BELBaR project itself.

This report will be updated at the end of the BELBaR project. It is anticipated that, when this report is updated, progress will have been made against these issues as a result of work undertaken in the BELBaR project itself – this will clearly demonstrate how the BELBaR project has been of significant benefit to the national programmes that are participating in it with regard to the treatment of colloids in performance assessment.

4.1 Finland

Issue	**Relevance to the Safety Case**	**Need for additional studies and link to BELBaR (ref to relevant work package)**
	Erosion	
Mechanisms of erosion of clay particles from the Bentonite surface	Loss of buffer mass potentially weakens the ability of buffer to limit the transport of groundwater solutes to and from the canister. The results of the safety analysis emphasize the importance of disposal canisters and other system parts in the immediate vicinity of canisters in limiting the release of radioactive substances. By understanding the mass loss rate limiting mechanisms in more details it might be possible to focus the future efforts to fewer aspects that provide sufficient resolution to the safety compromising issue of bentonite mass loss by erosion by dilute water.	Parameter sensitivity investigations by experimental means (with parallel numerical modelling) in geometrically, dynamically, chemically and possibly thermally conditions similar (enough) with respect to the ones in repository. That is WP2 and WP5.
Characteristics of Bentonite clay	Whereas monovalent cations have been considered even somewhat systematically in parametric investigations, divalent cations have not been studied that systematically. Should the	The role of divalent cations can be pursued in WP2 and WP4. Insoluble accessory minerals are a topic for a potential parallel (smallish) investigation in WP2.

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	existence and quantitative effect of divalent cations be argued, the importance of this currently one of the most outstanding uncertainties would become considerably lesser. Moreover, insoluble accessory minerals have been currently considered as having a potentially positive effect through forming a somewhat rigid weakly permeable layer terminating colloids release (i.e. a positive reserve FEP). The potential for and of a well-reasoned argumentation of the role of these minerals is currently uncertain.	
Groundwater Chemistry	See above regarding mono- and divalent cations and the related concentrations.	Monodivalent aspects have been pursued in previous investigations (see Figure 4) up to some extent, but it should be continued as proposed above.
Clay – Groundwater interactions	Changes in bentonite's porewater solute concentrations arising from diffusive mass transport and changes in void ratio (i.e. solid-to-liquid ratio) have been modelled. The related rates have been assumed to be limited by the availability of different porewater solutes and not by chemical kinetics. Mass loss rate can be assumed to have hydrodynamic contribution, but as indicated in the text immediately above Figure 4, the (geometrically similar but dynamically and chemically possibly dissimilar) downscaled tests have questioned the validity of numerical model. (Dissolving minerals are assumed to be exhausted they occur at all.)	Since the effects of divalent cations and insoluble accessory minerals can currently be investigated only by experimental means validation of experimental methods in conditions assessable numerically is needed. Similarity of conditions in downscaled tests with respect to the assumptions in the numerical model can be established as a joint effort of WP2, WP4 and WP5. Thereafter a validated argumentation for (the conditions for) maximum mass loss rate (with lesser uncertainties than earlier) to be used in safety case can be prepared. This is a cross-WP effort as well.
Groundwater velocity	Groundwater velocity has been considered as a variable. It has an evident contribution but its extent has been questioned and it is discussed above.	See above.
Clay extrusion paths	Since mass loss rate scales directly	Different apertures should be used

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	with fracture aperture, more extensive investigations to validate this assumption need to be committed. Fractures have been assumed to be planar with a constant aperture. Planning of variable aperture investigations have been initiated based on CFM experiences.	in WP2. Variable aperture tests should be discussed and possibly initiated within WP2.
Colloid, radionuclide and host rock interaction		
Colloid mobility controlling processes	Clay colloids have not been considered radionuclide carriers due the assumed low contribution.	WP3 could either validate or invalidate this assumption i.e. explicitly revise this assumption.
Retention processes	See above.	See above.
Radionuclide sorption	See above.	See above.
Colloid Stability		
Colloid stability controlling processes	Stability of compacted bentonite in dilute porewater conditions has been evaluated by laboratory measurements. In the context of KBS-3 the (or should it be "a"?) controlling process is hydration of exchangable cations limited by the availability of cation free water. The extent of this hydration depends on the characteristics of cations. The edge-to-face interactions are not taken into account. When the shear force induced by flowing groundwater exceeds gel's/sol's cohesive forces mass loss occurs more easily. Currently the uncertainties in geochemical conditions are greater than in uncertainties in the stability limit.	No new needs.
Influence of other factors to colloid stability	Only the effect of potentially formed stable bed of accessory minerals. Accessory minerals seem to enrich near the bentonite-groundwater interface but its stability has not been shown that is essential.	See proposal on "*Characteristics of bentonite clay*".
Conceptual and mathematical models		
Current model(s): Erosion of the Bentonite buffer	The factors considered are • groundwater velocity • fracture aperture	See proposal on "*Clay-groundwater interaction*".

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	• transport resistance of bentonite gel in terms of diffusivity, *D* (lumping various factors) • gel cohesivity in terms of viscosity, □ (lumping various factors) Small-scale tests suggests though groundwater ionic strength and the presence of divalent cations being the dominant factors. With respect to these factors pessimistic assumption neglecting safety promoting aspects have been used.	
Current model(s): Radionuclide transport mediated by Bentonite colloids	Clay colloids have not been considered radionuclide carriers in Posiva's safety case due the assumed low contribution.	This assumption will be considered (according to the original plans) in terms of reversibility of sorption in WP5 and WP3.

4.2 Sweden

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	Erosion	
Mechanisms of erosion of clay particles from the Bentonite surface	The mechanisms of erosion of clay particles from the bentonite surface have a strong and direct impact on the Safety Case for a KBS-3 repository in Sweden. With the current model erosion will cause a loss of buffer performance under some conditions. In turn, this may lead to corrosion failures of the canisters. Since the corrosion failure has the biggest impact on risk in the SR-Site assessment, less pessimistic approach may have a significant impact on the calculated risk.	The mechanisms of clay colloid release are not fully understood. This will be studied in WP2. The quantitative models can be improved with new data. This will be done in WP5
Characteristics of Bentonite clay	The current model is based on an “idealized” pure sodium-montmorillonite. Accessory minerals are supposed to be washed out. Even pure sodium-montmorillonites behaves differently with respect to erosion. A better understanding of the details could	The stability of different bentonites will be evaluated in WP4

Issue	**Relevance to the Safety Case**	**Need for additional studies and link to BELBaR (ref to relevant work package)**
	improve the models, but possibly also guide material selection.	
Groundwater Chemistry	The key factor for colloid stability is the ionic strength and the content of divalent cations. pH should have an effect, but the pH-range considered in the safety case is rather limited	The effect of mixed monovalent/divalent systems is still one of fundamental uncertainties. This issue will be studied in WP2, WP4 and WP5
Clay – Groundwater interactions	The buffer and the groundwater will never reach a true equilibrium. Observations from small scale experiments can therefore not be directly used in the safety case.	WP5 and WP1 will discuss how the studied processes should be integrated in the safety case.
Groundwater velocity	According to the assessment model in SR-Site, the loss of bentonite is affected by the groundwater velocity (to the 0.41 power)	It is important to verify the dependence between the groundwater velocity and the erosion rate. This will be done in WP2
Clay extrusion paths	Extrusion of clay into a fracture is an integral part of the current model and will have a strong impact on the mass loss	The effect of fracture geometry is studied in WP2
Colloid, radionuclide and host rock interaction		
Colloid mobility controlling processes	Rather than attempting to develop detailed process models for colloid-facilitated transport, potential mitigating processes are ignored so as to place an upper bound on the possible effect.	WP3 will study possible retardation mechanisms
Retention processes	See above	See above
Radionuclide sorption	To assess the possible role of rapid reversible sorption/desorption onto colloids in facilitating transport, the following assumptions are adopted: (i) equilibrium sorption of radionuclides onto mobile and immobile colloids, (ii) equilibrium sorption of colloids onto fracture surfaces, and (iii) colloid-free matrix pore space (conservative assumption, but also realistic for the small pore sizes of granitic rock). Although not strictly needed, an additional assumption of constant (in space) colloid concentration is used, consistent with the modelling cases of this report that involve colloid-facilitated transport.	Reversibly is assumed and this needs to be varied in WP3
Colloid Stability		
Colloid stability controlling processes	The colloid stability criterion in SR-Site was based only on laboratory	All aspects of colloid stability will be studied in WP3

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	observations and considers only total charge of the groundwater. The value may have a strong impact on the risk. An order of magnitude change in any direction would have a significant impact.	
Influence of other factors to colloid stability	Filtration has been discussed as a possible mean to reduce erosion. It is however difficult to prove that the process is efficient.	It is somewhat unclear how much effort will be put into this issue within BELBaR
Conceptual and mathematical models		
Current model(s): Erosion of the Bentonite buffer	The montmorillonite release rate, $R_{Erosion}$, is found to be proportional to the water velocity, v, to the power 0.41 and directly proportional to the aperture, δ. The validity of the model is discussed in [1].	Parameters, model improvements and verification are discussed under the "Erosion" heading above. Model improvement and testing will be done in WP5
Current model(s): Radionuclide transport mediated by Bentonite colloids	The effective transport parameters described under "radionuclide sorption" above these have been incorporated into the MARFA code [42]	The issue is if the assumption of reversible sorption is correct. If so, the impact of radionuclide transport with colloids in a KBS-3 repository in Forsmark will be insignificant

4.3 United Kingdom

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
Erosion		
Mechanisms of erosion of clay particles from the Bentonite surface	Erosion will cause a loss of bentonite buffer performance under some conditions. This may lead to corrosion failures of the canisters. Since corrosion failure leads to the largest impact on risk in post-closure assessments, a less pessimistic approach may have significant impacts on the calculated risk.	The mechanisms of clay colloid release are not fully understood. This will be studied in WP2. The quantitative models can be improved with new data. This will be done in WP5.
Characteristics of Bentonite clay	The bentonite is assumed to act as a colloid filter.	The stability of different bentonites will be evaluated in WP4
Groundwater Chemistry	The key factor for colloid stability is the ionic strength and the content of divalent cations. pH should have an effect, but the pH-range considered	This issue will be studied in WP2, WP4 and WP5.

Issue	**Relevance to the Safety Case**	**Need for additional studies and link to BELBaR (ref to relevant work package)**
	in the safety case is likely to be limited. The GDF site is currently unknown so no site-specific groundwater chemistry data are available for the UK.	
Clay – Groundwater interactions	It is unlikely that the buffer and the groundwater will ever reach a true equilibrium. This implies that observations from small scale experiments cannot be directly used in the safety case.	WP5 and WP1 will discuss how the studied processes should be integrated in the safety case.
Groundwater velocity	The loss of bentonite will be affected by the groundwater velocity and it is important to verify this dependence and erosion rates.	This will be done in WP2
Clay extrusion paths	Piping will take place if the water pressure in a fracture is higher than the total pressure in the clay and the clay's shear resistance, the hydraulic conductivity of the clay is low enough that water flow into the clay ceases to counteract the water pressure in the fracture. Piping occurs before full saturation of the buffer. Extrusion of clay into a fracture will have a strong impact on the mass loss.	The effect of fracture geometry will be studied in WP2.
Colloid, radionuclide and host rock interaction		
Colloid mobility controlling processes	Colloidal clay particles may attach strongly to gas bubbles (flotation) and their transport may be either accelerated or retarded compared to water flow depending upon bubble transport [43]. There is little information available concerning the transport of colloids by flotation in geological systems.	WP3 will study possible retardation mechanisms.
Retention processes	Retardation of colloid transport in the far field, by sorption of colloids onto rock surfaces, will delay the arrival of radionuclides in the biosphere. The extent of this isn't	See above

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	currently taken into account.	
Radionuclide sorption	Reversible, linear sorption of radionuclides onto colloids has been assumed. For the case of reversible radionuclide sorption to colloids and reversible colloid surface attachment, colloid retardation will only have a significant impact on the retardation of radionuclides if there is significant partitioning of radionuclides to the colloidal phase.	Reversibility is assumed and this needs to be verified in WP3.
Colloid Stability		
Colloid stability controlling processes	Colloid stability studies have found that model colloids that possess a significant net negative charge at neutral pH, i.e. silica and illite clay, show the greatest stability under neutral pH conditions [28].The principal mechanism of inorganic colloid stabilisation against aggregation is electrostatic repulsion. Clay particles possess a structural negative charge due to isomorphous substitution in which silicon atoms may be replaced by aluminium atoms, for example. To preserve electroneutrality, the particle is surrounded by a diffuse atmosphere of counterions. Colloidal particles are thermodynamically metastable with respect to bulk solid phases owing to their higher surface free energy, which increases with decreasing particle size. As a result, smaller particles will tend to dissolve while larger particles or solid phases will grow preferentially [28].	All aspects of colloid stability will be studied in WP3.
Influence of other factors to colloid stability	Additive effects aren't currently considered and little is known about colloid behaviour in the presence of microbes or various complexing agents (for example; do	See above. Unsure how much focus there will be on other influences as part of BELBaR.

Issue	Relevance to the Safety Case	Need for additional studies and link to BELBaR (ref to relevant work package)
	radionuclides sorb preferentially to cellulose degradation products?) Colloid size, solution ionic strength and water flow rate are factors which strongly influence colloid migration. Association of inorganic particles with natural organic compounds is an important mechanism for colloid stabilisation in surface and near-surface waters, where concentrations of natural organic materials may be high. Due to the large amount of organic materials in the UK waste inventory, this mechanism could potentially operate to stabilise and enhance colloid populations in the near-field porewater. Limited experimental evidence to date does not support this contention, however, although this remains an area of uncertainty [26].	
Conceptual and mathematical models		
Current model(s): erosion of the bentonite buffer	The current model is discussed in Section 3.3.3.	Model improvement and testing will be done in WP5.
Current model(s): Radionuclide transport mediated by Bentonite colloids	At the colloid concentrations likely in the far field, a significant increase in risk could arise if a proportion of the radionuclides associated with colloids are irreversibly sorbed. In that case the risk will depend on the mobility and transport properties of colloids through the geosphere and the timescale over which individual particles remain dispersed in the fluid (particle lifetimes).	The issue is whether the assumption of reversible sorption is correct. If so, the impact of radionuclide transport with colloids in a GDF will be insignificant.

5 Summary

This report is a key early-project deliverable for the EC BELBaR project, and considers the current state-of-the-art treatment of colloids issues in PA for a range of participating national radioactive waste management programmes.

The report provides information on the structure of the BELBaR project, noting how the five constituent Work Packages are inter-related and designed to provide an information flow throughout the project. This relationship between the Work Packages is intended to ensure, for example, that the development of understanding of colloids processes at a research level is reflected appropriately in the treatment of colloids in performance assessment, and that the research studies themselves are focussed at the outset on key performance assessment issues – this is an iterative process, necessitating the close collaboration of a range of scientific disciplines and the undertaking of needs-directed research activities.

The report has achieved the following:

- The current treatments of colloid issues in PA (on the basis of national organisations involved in BELBaR only) have been described;
- The limitation of previous studies and uncertainties related to colloids have been noted;
- The needs for additional studies of colloids issues, and their PA relevance, have been discussed ;
- Such additional studies have been linked to planned work in the BELBaR project, and the relevance of and expected benefit from BELBaR WPs 2-5 have been identified.

Each of the three national waste management organisations represented in BELBaR Work Package 1 has produced a description of how it currently considers colloids in performance assessment – the current state-of-the-art.

This information has subsequently been used to identify current issues affecting the treatment of colloids in performance assessment in national programmes. These issues are identified in a series of tables.

The issues are linked in these tables as appropriate to the Work Packages considered in the BELBaR project, clarifying how the issues raised by national programmes regarding the treatment of colloids in performance assessment will be progressed by the BELBaR project itself. Note that the relevance of the information contained in these tables to national programmes varies on a programme by programme basis.

Through the work now being undertaken in the BELBaR project, progress will be made against these issues. This report will be updated at the end of the BELBaR project to reflect this progress – the revised version of the report will clearly demonstrate how the BELBaR project has been of significant benefit to the national programmes that are participating in it with regard to the treatment of colloids in performance assessment.

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