BELBaR: *Investigation of erosion processes in bentonite engineered barriers systems of a repository in crystalline rock and their impact on the long-term performance of the repository*

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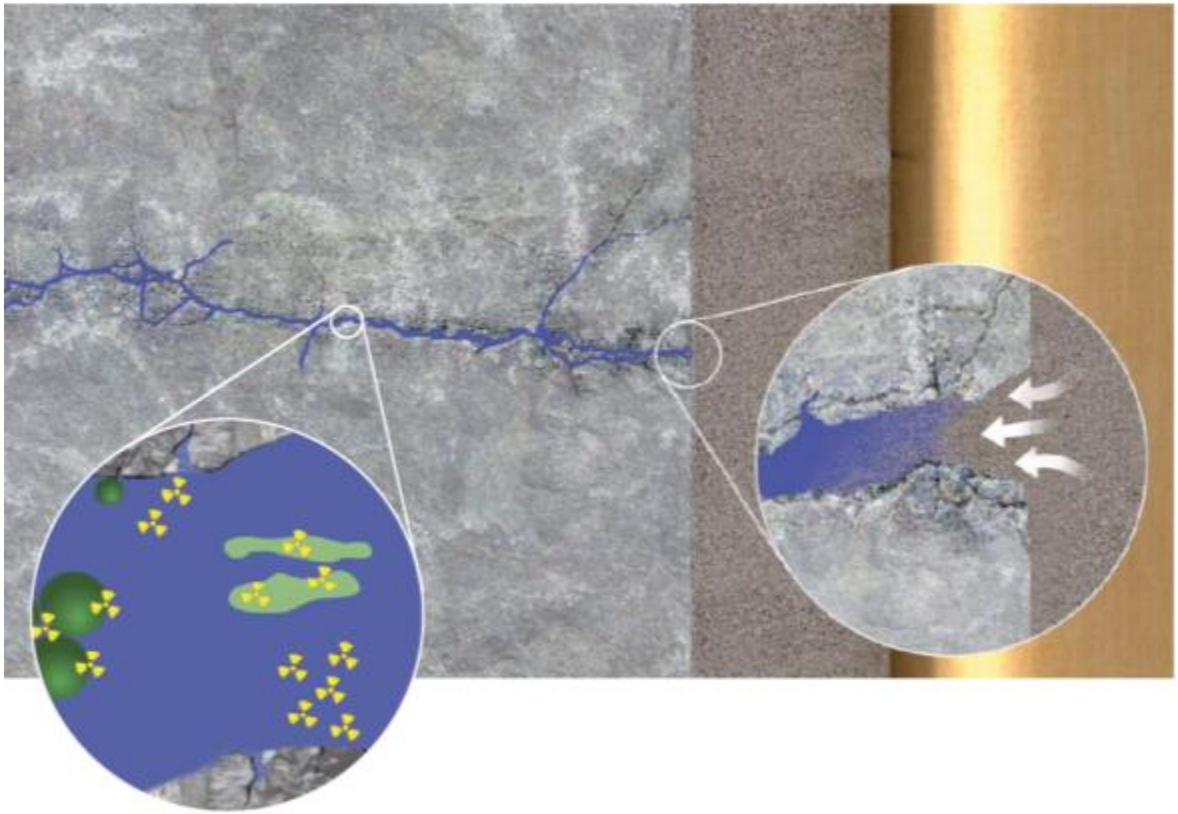
BELBaR

- BELBaR was a Collaborative Project within the Seventh Framework Programme of the European Atomic Energy Community (Euratom) for nuclear research and training activities.
- The main aim of BELBaR was to increase knowledge of the processes that control clay colloid stability, generation and its ability to transport radionuclides.
- The overall purpose of the project was to come up with a new way of treating issues in long-term safety/performance assessment.
- The project started March 1, 2012 and had a duration of 48 months.
  - BELBaR endedFebruary 28, 2016
- The project had 14 partners from seven European countries

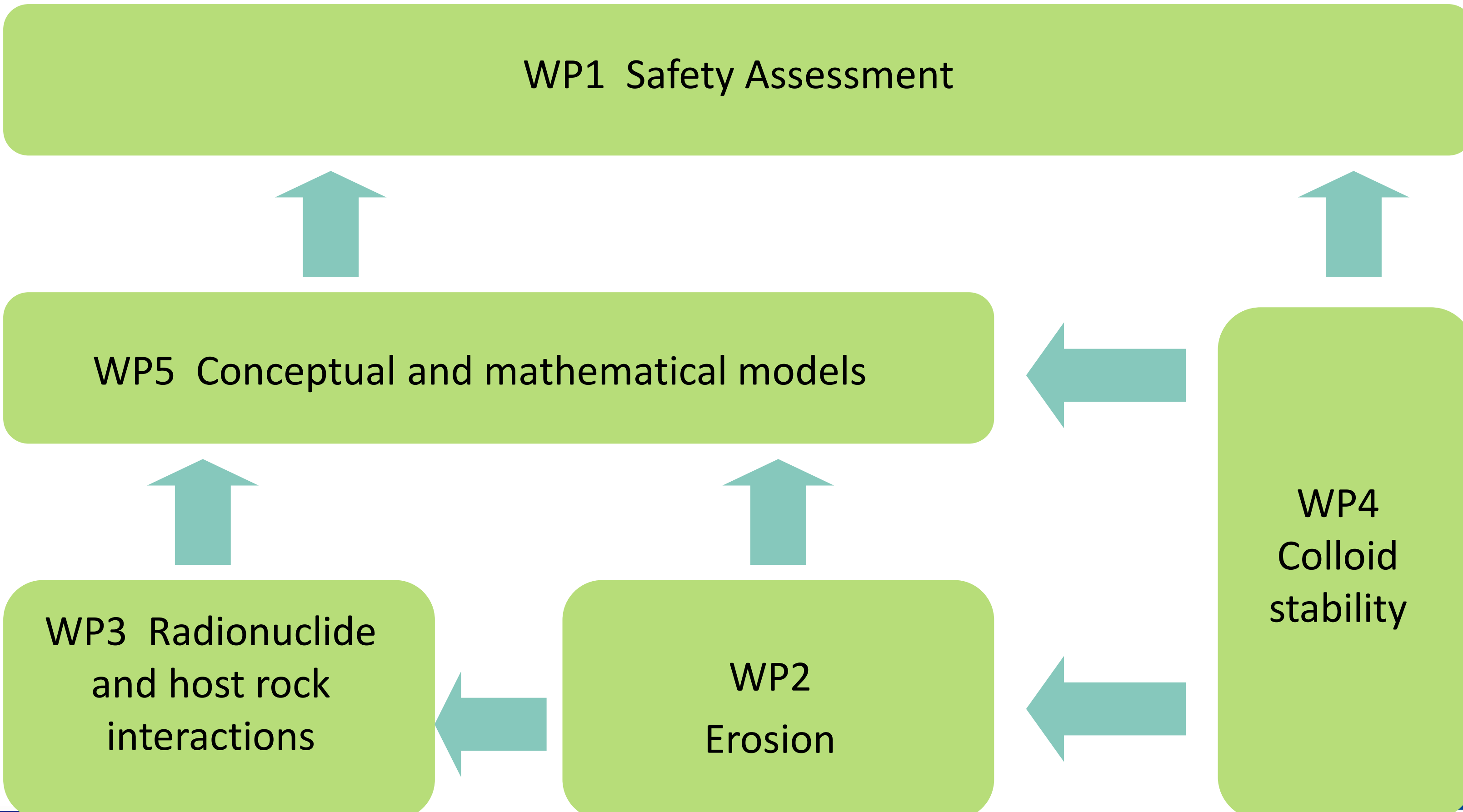
Partners

- SKB: Svensk Kärnbränslehantering, Sweden
- CIEMAT: Centro de Investigaciones, Energeticas, Medioambientales y Technologicas Spain
- NRI: Nuclear Research institute Rez plc, Czech Republic
- KIT: Karlsruhe Institut of Technology, Germany
- Posiva OY, Finland
- VTT:Technical Research Instuitute of Finland
- Clay Technology AB, Sweden
- University of Jyväskylä, Finland
- KTH: Kungliga Tekniska Högskolan, Sweden
- NDA: Nuclear Decommissioning Authority, United Kingdom
- B+Tech Oy, Finland
- University of Manchester ,United Kingdom
- Helsinki University, Finland
- Lomonosov Moscow State University, Russia

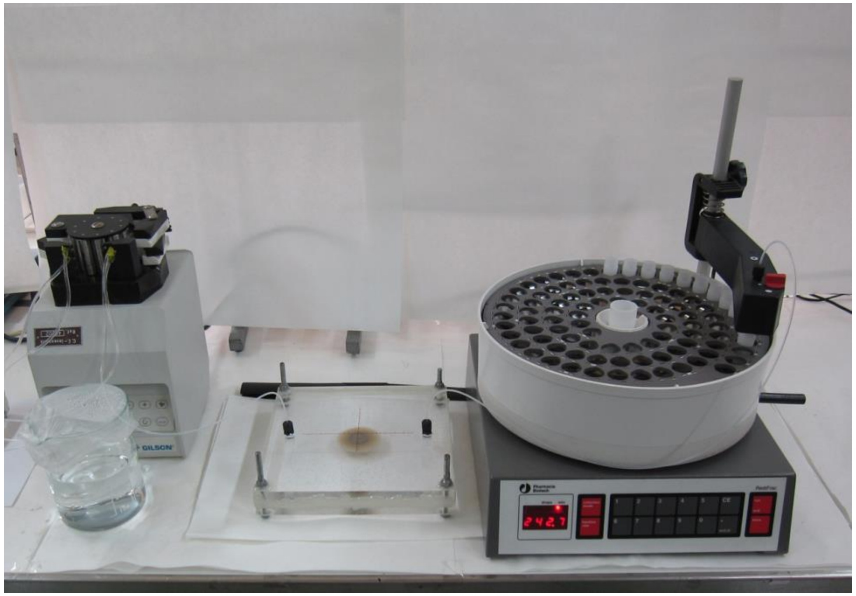
The problem to study

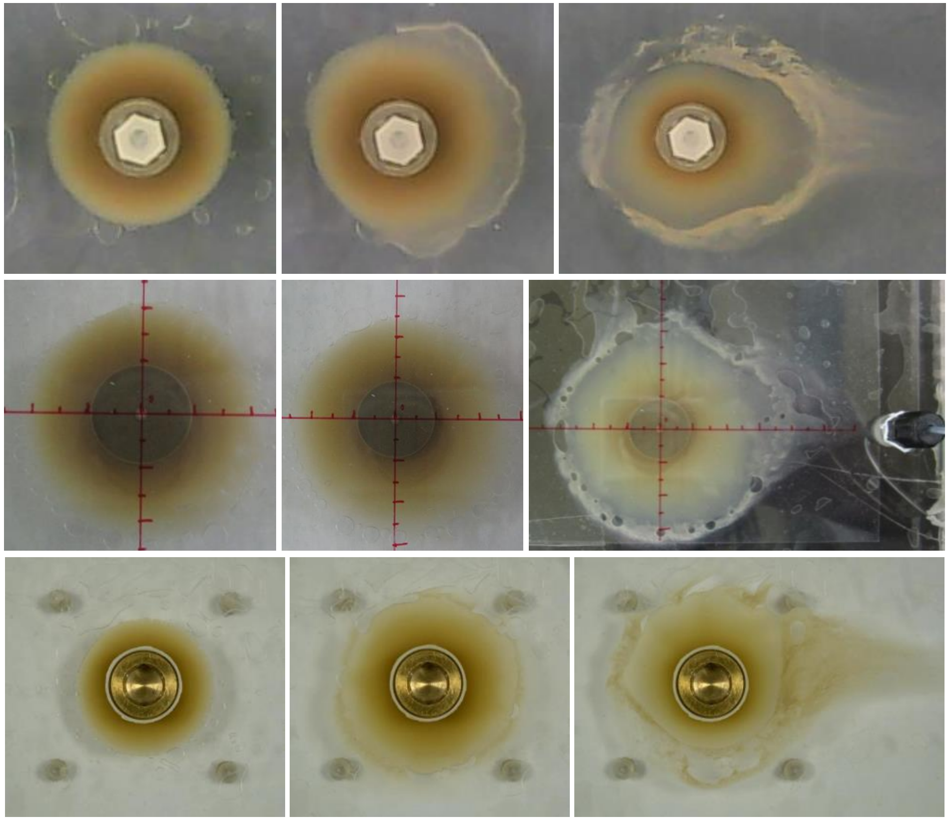


BELBaR Project WP Linkages: Information flow



WP2: Erosion





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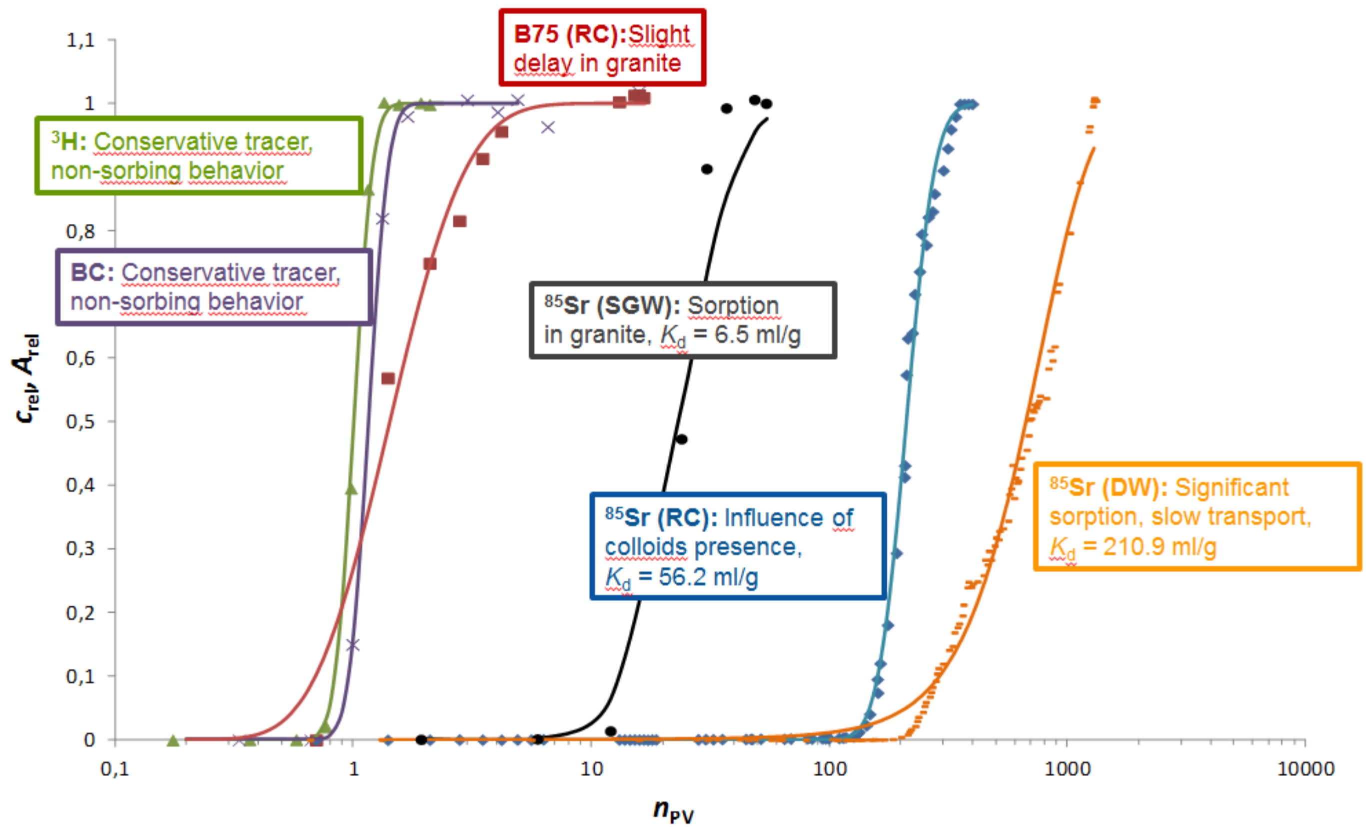
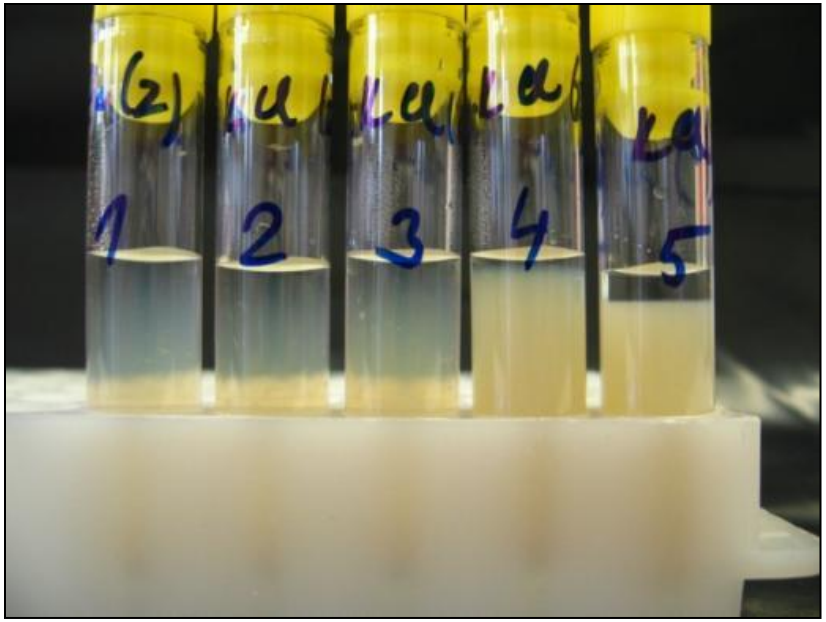
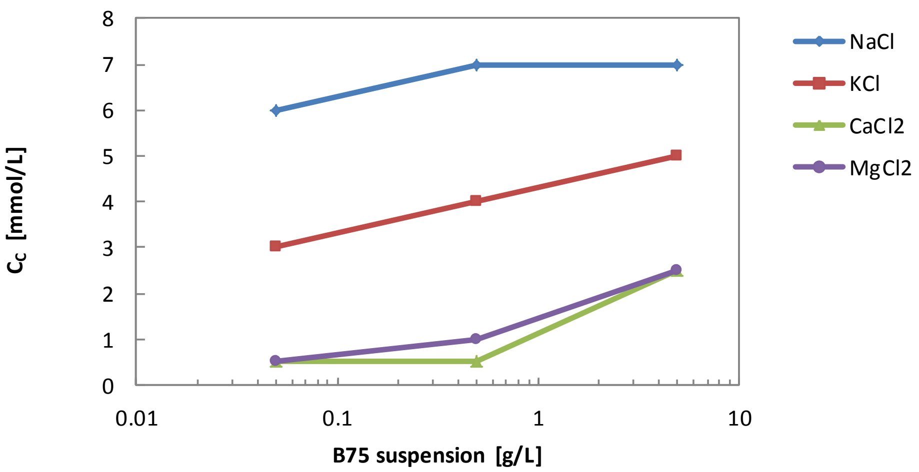
WP3: Radionuclide and host rock interactions



WP4 Colloid stability





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WP5 Conceptual and mathematical models

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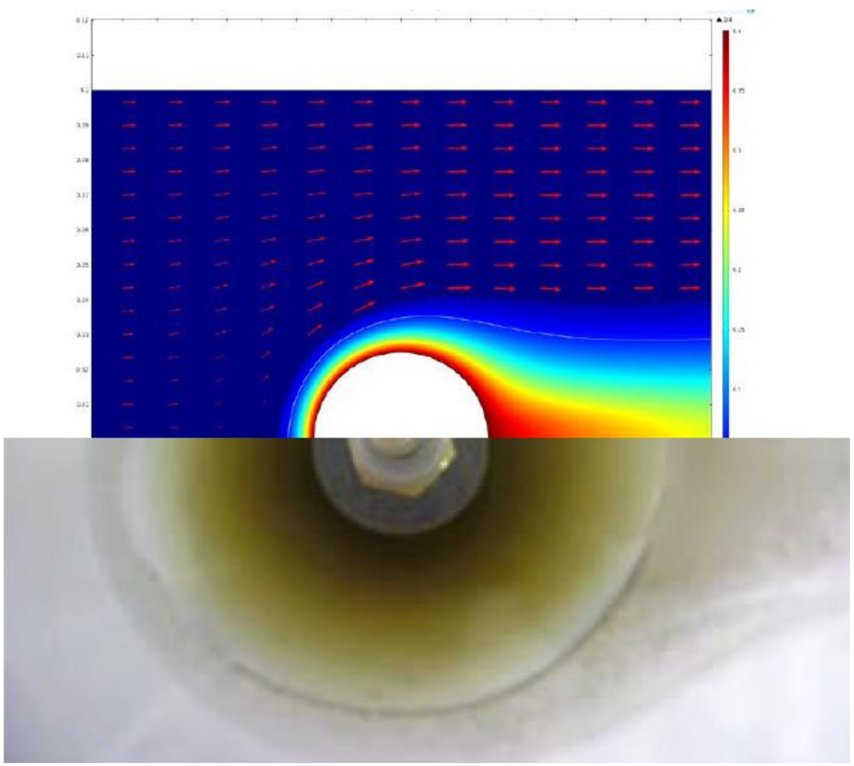

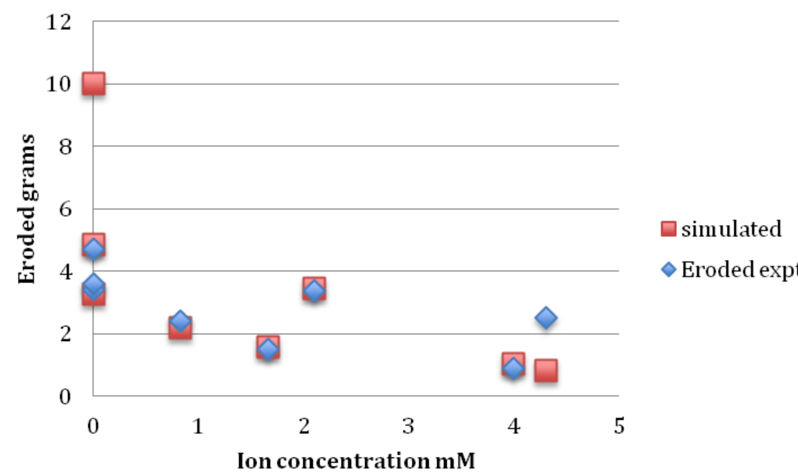

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Synthesis of issues: Erosion

Mechanisms of erosion of clay particles from the bentonite surface

Safety case position at start of BELBaR	Outcomes of BELBaR
Erosion will cause a loss of bentonite buffer performance under some conditions. This may lead to corrosion failures of the canisters. Corrosion failure leads to the largest impact on risk, a less pessimistic approach may have significant impacts on the calculated risk.	In general, erosion/ colloid generation is rapid initially, but decreases with time and in some cases stops altogether. In static experiments – equilibrium is reached – maximum quantity of colloids generated depends on initial conditions – but erosion will not be continuous. Chemical forces driving dispersion processes are considered to be more important than mechanical forces even in the dynamic system Potential connection between flow rate and erosion when ionic strength of GW is below the CCC

Synthesis of issues: Erosion

Characteristics of Bentonite Clay

Safety case position at start of BELBaR	Outcomes of BELBaR
Divalent cations have not been studied that systematically. Should the existence and quantitative effect of divalent cations be argued, the importance of this outstanding uncertainty would reduce.	Compaction density of bentonite clay – with higher compaction, the quantity of particles produced is observed to be higher. Higher erosion observed in Na- exchanged bentonites. Low (and sometimes no) erosion observed in Ca- exchanged bentonites – even in low ionic strength conditions. A small proportion of Na in bentonite (25%) is enough to promote significant dispersivity of the clay. Link between maximum eroded mass and the smectite content of the clay

Synthesis of issues: Erosion

Groundwater Chemistry

Safety case position at start of BELBaR	Outcomes of BELBaR
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.	Ionic strength of GW is an important factor with respect to erosion. In general it has been observed that increasing ionic strength, decreases colloid generation. Divalent cations (Ca^{2+}) are more effective as coagulants than monovalent cations (Na^{+}). Presence of Ca^{2+} in solution inhibits the formation of colloids. Erosion is only observed at very dilute conditions. Question of conservatism with respect to consideration of the deionised GW scenarios – but consideration needs to be given to composition of glacial melt waters

Synthesis of issues: Erosion

Clay- Groundwater interactions

Safety case position at start of BELBaR	Outcomes of BELBaR
Changes in bentonite porewater solute concentrations can be modelled. The related rates assumed to be limited by the availability of different porewater solutes. Mass loss rate assumed to have hydrodynamic contribution. The buffer and the groundwater never reach a true equilibrium.	Groundwater evolution in the long term will affect the chemistry of the clay/ groundwater system. Unlikely that mechanical shear is the key mechanism to perturb gel phase. Hysteresis effect could enable clay to be stable to erosion. Calcium incorporation in an open/ dynamic system could reduce/ eliminate the production of colloids.

Synthesis of issues: Erosion

Groundwater velocity

Safety case position at start of BELBaR	Outcomes of BELBaR
Groundwater velocity has been considered as a variable. The loss of bentonite will be affected by the groundwater velocity and it is important to verify this dependence for erosion rates.	Understanding the effect of the presence of a hydraulically active fracture – does water flow at the bentonite surface or gel- front increase colloid generation. In tests where erosion was observed erosion is well correlated to GW velocity. Tests conducted in less dilute conditions saw less mass loss, therefore potentially the use of maximum erosion rates to estimate mass loss could lead to overly conservative erosion predictions. Observed that system chemistry is more relevant than flow velocity in terms of driving erosion processes.

Synthesis of issues: Erosion

Clay extrusion paths

Safety case position at start of BELBaR	Outcomes of BELBaR
Fractures have been assumed to be planar with a constant aperture. Extrusion of clay into a fracture is an integral part of the current model and will have a strong impact on the mass loss. Piping may occur before full saturation of the buffer under certain circumstances.	Horizontal and sloped fractures display a different mechanism of mass loss. Where all other test conditions are identical – increased slope angle leads to increased mass loss. However, effect of slope is more dramatic at lower angles (0˚– 25˚) compared to 45˚-90˚. Irrespective of slope at a cation charge greater than or equal to 8.6 mEq – rate of erosion is effectively zero.

Synthesis of issues: Colloid, RN & host rock interaction

Retention processes

Safety case position at start of BELBaR	Outcomes of BELBaR
Retardation of colloid transport in the far field, will delay the arrival of radionuclides in the biosphere. The extent of this isn't currently taken into account.	Retention of colloids has been observed under conditions which would have been through to be unfavourable. Rock matrix diffusion is cited in terms of an interpretation of previous observations for the behaviour of colloids in flow paths. In terms of colloid diffusion coefficients – the main differences observed are attributed to size variation.

Synthesis of issues: Colloid, RN & host rock interaction

Radionuclide sorption

Safety case position at start of BELBaR	Outcomes of BELBaR
To assess the possible role of rapid reversible sorption/desorption onto colloids in facilitating transport, the following assumptions have been adopted: 1. equilibrium sorption of radionuclides onto mobile and immobile colloids, 2. equilibrium sorption of colloids onto fracture surfaces, and 3. colloid-free matrix pore space (conservative assumption, but also realistic for the small pore sizes of granitic rock). Reversible, linear sorption of radionuclides onto colloids has been assumed.	Sorption reversibility of irreversibility in terms of the migration of radionuclides can potentially be limiting or supporting. Its been observed that the affinity of radionuclides towards the rock was in general greater than the affinity towards the colloids, indicating reversible sorption. Colloid mediated transport of radionuclides is dependant on the composition of the liquid phase – link to colloid stability and competition for sorption sites between ions and radionuclides can speed up transport. Full reversibility of sorption cannot be guaranteed, reversibility kinetics depend on geochemical parameters.

Synthesis of issues: Colloid stability

Colloid stability controlling processes

Safety case position at start of BELBaR	Outcomes of BELBaR
Stability of compacted bentonite in dilute porewater conditions has been evaluated by laboratory measurements. The controlling process is hydration of exchangable cations limited by the availability of cation free water. Currently the uncertainties in geochemical conditions are greater than in uncertainties in the stability limit. 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.	Provided that the ionic strength of groundwater is above the CCC, highly compacted clays in engineered barriers will act as a swelling repulsive paste and expand. Colloid stability is influenced by the differing mineralogy of different bentonites Apparent hysteresis effect where aged gels are observed to be stronger.

Synthesis of issues: Colloid stability

Influence of other factors on colloid stability

Safety case position at start of BELBaR	Outcomes of BELBaR
Accessory minerals seem to enrich near the bentonite-groundwater interface. Filtration has been discussed as a possible mean to reduce erosion. 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. This mechanism could potentially operate to stabilise and enhance colloid populations in the near-field porewater, this remains an area of uncertainty.	Na and K (M^{+}) and Mg and Ca (M^{2+}) act in a similar way during coagulation process Interaction of smectite with a mineral such as kaolinite or Al2O3 produces aggregation of particles. Organic matter is able to stabilise colloids in NaCl electrolyte. In $MgCl_2$ and $CaCl_2$, clay colloids undergo fast coagulation independently of the presence of organic matter.

Synthesis of issues: Conceptual and mathematical models

Erosion of the bentonite buffer and radionuclide transport mediated by bentonite colloids

Safety case position at start of BELBaR	Outcomes of BELBaR
The factors considered are; 1. groundwater velocity 2. fracture aperture 3. transport resistance of bentonite gel in terms of diffusivity 4. gel cohesivity in terms of viscosity Pessimistic assumption neglecting safety promoting aspects have been used. 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 particle lifetimes.	Based on the experimental evidence observed, review our conceptual and mathematical models of the influence of colloids on the erosion of the bentonite buffer

Summary

The key issues addressed in the final State-of-the-art report from each WP:

- Bentonite erosion and production of colloids (WP2) – understanding of the main mechanisms of erosion of clay particles from the bentonite surface and quantification of the (maximum) extent of the possible bentonite erosion under different physio-chemical conditions.
- Colloid, radionuclide and host rock interactions (WP3) –determined the sorption reversibility of radionuclides to the colloids and whether the current assumption of reversible sorption can be justified.
- Clay colloid stability (WP4) – better understanding and representation of the stability and mobility of clay colloids under repository site conditions.
- Improvements to models (WP5) –obtained improved, validated models of colloid influence on the erosion of the bentonite buffer and colloid-mediated radionuclide transport.

Overall, moving towards a justification for a review of current pessimistic assumptions in the safety case regarding colloid behaviour, to present a more realistic and confident safety case.

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