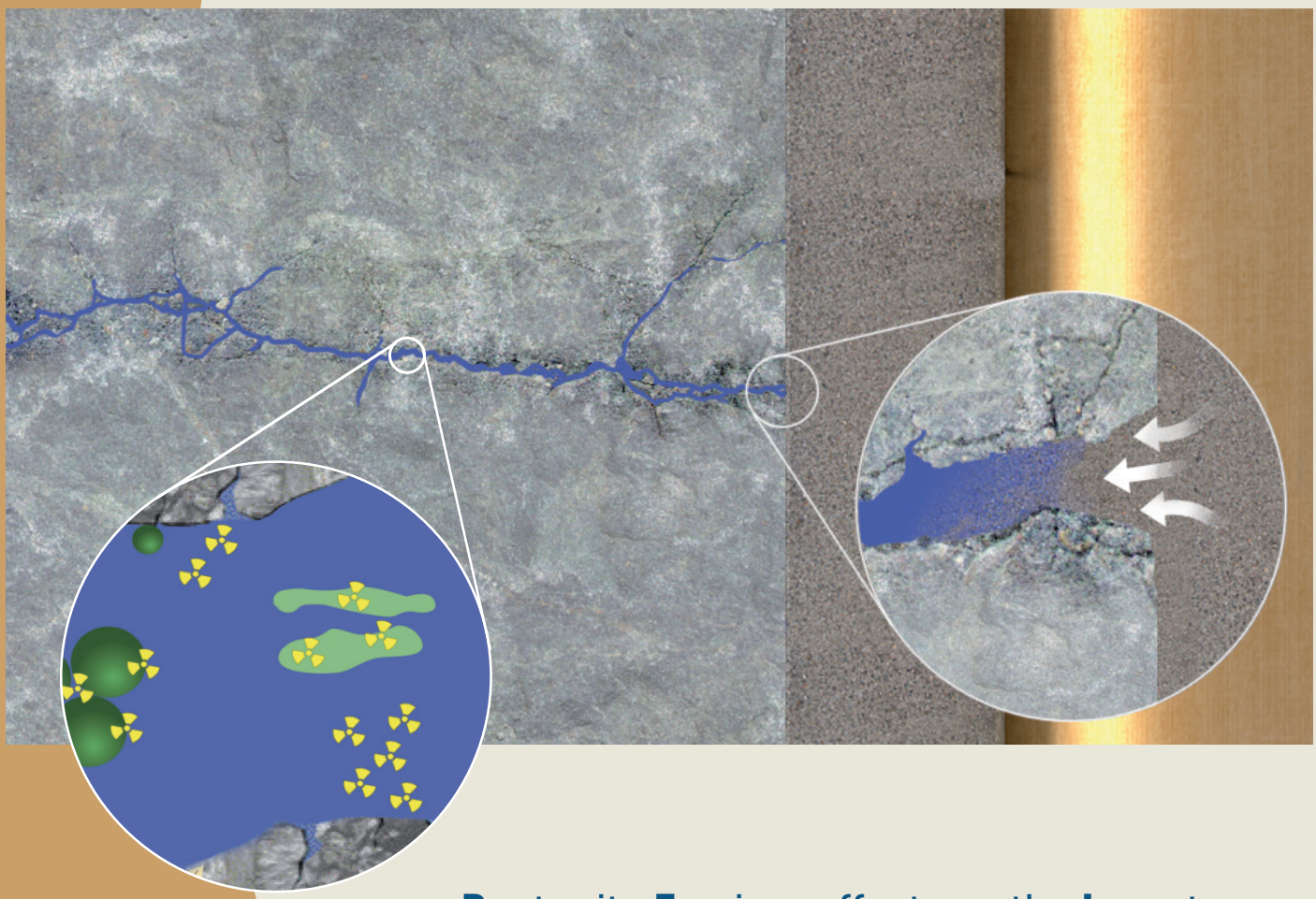
The BELBaR project has received funding from the European Atomic Energy Community's (EURATOM) 7th Framework Programme (FP7/2007-2011) under the grant agreement No. 295487

Summary of the BELBaR project



Bentonite **E**rosion: effects on the **L**ong term performance of the engineered **B**arrier **a**nd **R**adionuclide transport

Start date 01/03/2012
End date 29/02/2016

BELBaR

The BELBaR project was a collaborative project based on the desire to improve the long-term safety assessments for geological disposal facility concepts for spent fuel/ high level waste that combine a clay engineered barrier system (EBS) with a fractured rock. BELBaR partners included national radioactive waste management organisations (WMOs) from a number of countries, research institutes, universities and commercial organisations working in the radioactive waste disposal field.
The formation and stability of colloids from the EBS may have a direct impact on assessed risk from a geological disposal facility in two aspects;

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

This is illustrated in Figure 1

An increased understanding of the processes involved in the above aspects will have a significant impact on the outcome of future assessments. Therefore, the main aim of the BELBaR project was to increase the knowledge of the processes that control clay colloid generation, stability and their ability to transport radionuclides and reduce the uncertainties in the description of the effect of clay colloids in long-term performance assessments. To meet this aim, a number of experimental and modelling activities were undertaken within the project. The scientific part of the project was structured into five separate work packages dealing with different aspects of the issues.

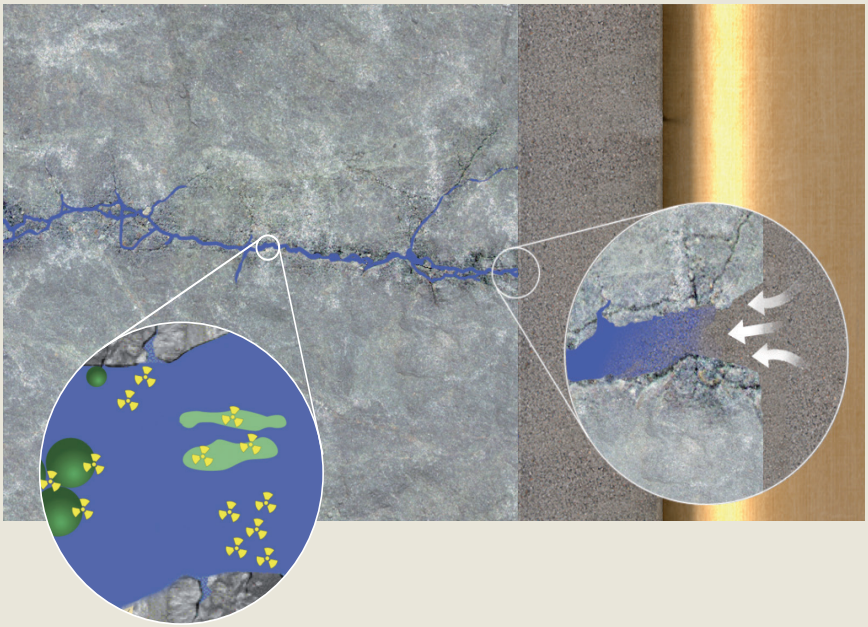


Figure 1 A schematic illustration of colloid generation from a bentonite EBS (right circle) and colloid transport of radionuclides (left)

WP1 Safety assessment

The objectives of WP1 at the outset of the BELBaR project were to identify and synthesise the current treatment of the relevant processes in existing safety assessments. This review was used to inform the direction of the experimental and modelling programme undertaken within the framework of the BELBaR project.
The decision tree shown in Figure 2 can help to form a basis of assessing the potential importance of colloids. By answering a series of questions concerning possible colloid behaviour and considering the underpinning evidence, this can provide a framework for how colloids are treated in a particular safety assessment.
At the end of the BELBaR project, the objective of WP1 was, by drawing on the results obtained within the respective work packages, to consider, and where appropriate, to recommend opportunities for where WMOs could review how colloids and related phenomena are considered in the long-term safety case.

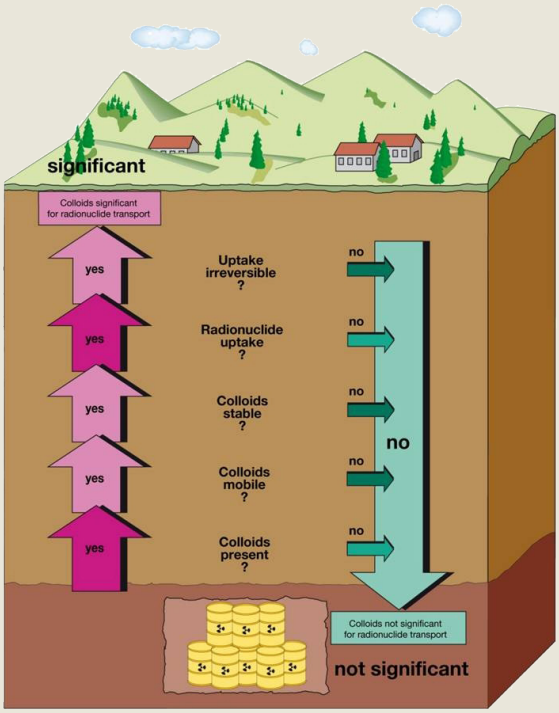
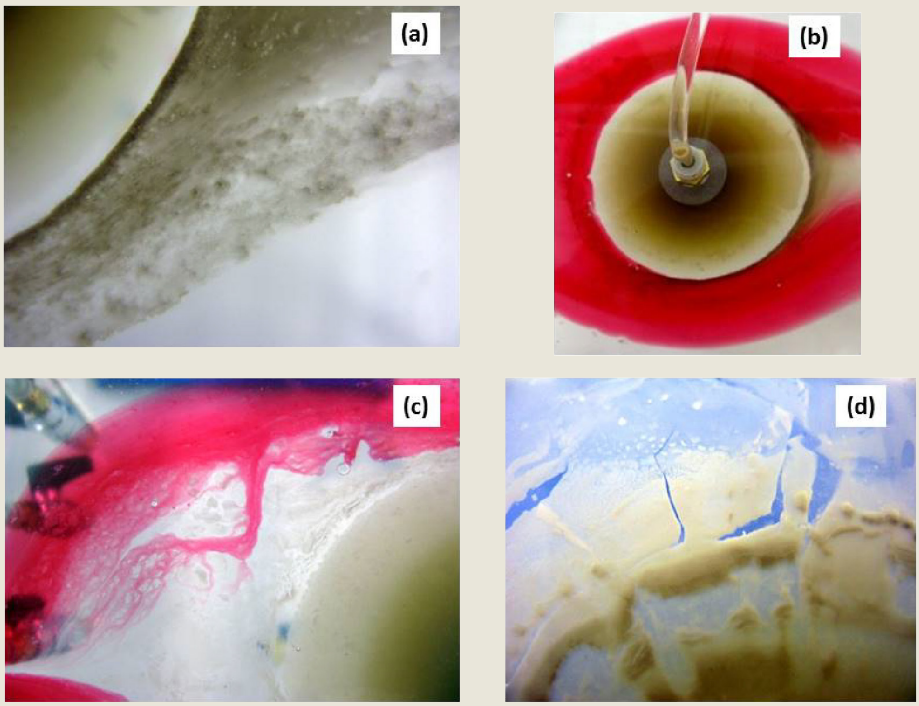


Figure 2 Basis for assessing the potential importance of colloids in performance assessment.

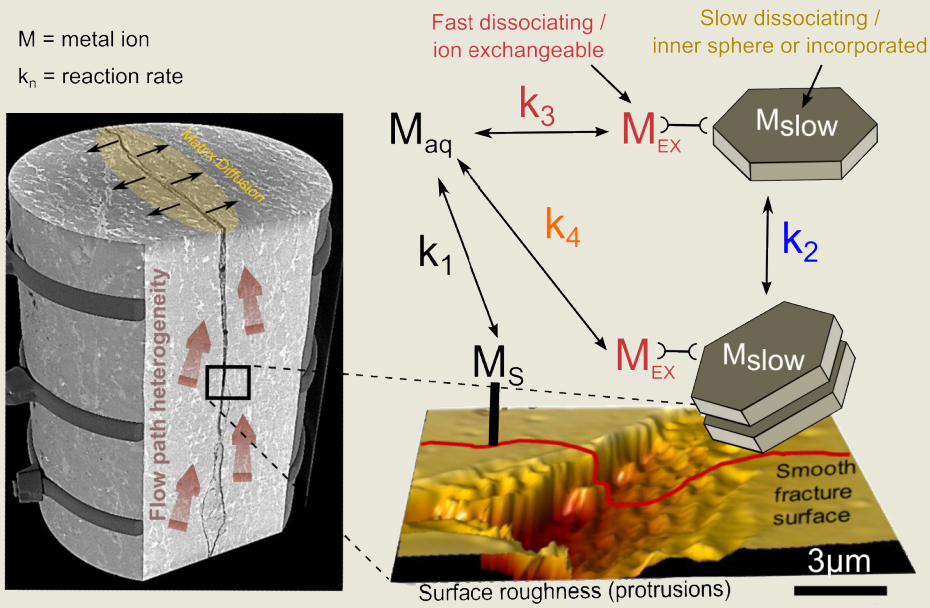
WP2: Bentonite erosion



The main objective of WP2 was 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.
Figure 3 shows examples of experimental observations of bentonite erosion and attenuation of erosion under different experimental conditions.

Figure 3 Photographic images showing a) the interface between the inner zone of extruded material and the outer zone of eroding material for a test with sodium montmorillonite against deionized water at 456 h, b) flow visualization of the same test highlighting the permeability of the eroding material zone and the relative impermeability of the extruded material zone, c) flow visualization of a test with 50/50 calcium/sodium montmorillonite against 4 mM NaCl indicating the semi-permeability of the eroding material zone and d) the extrusion/erosion interface from a test with 50/50 calcium/sodium montmorillonite against double strength Grimsel groundwater simulant demonstrating that the eroding material forms into extended sheets of rigid, coherent material.

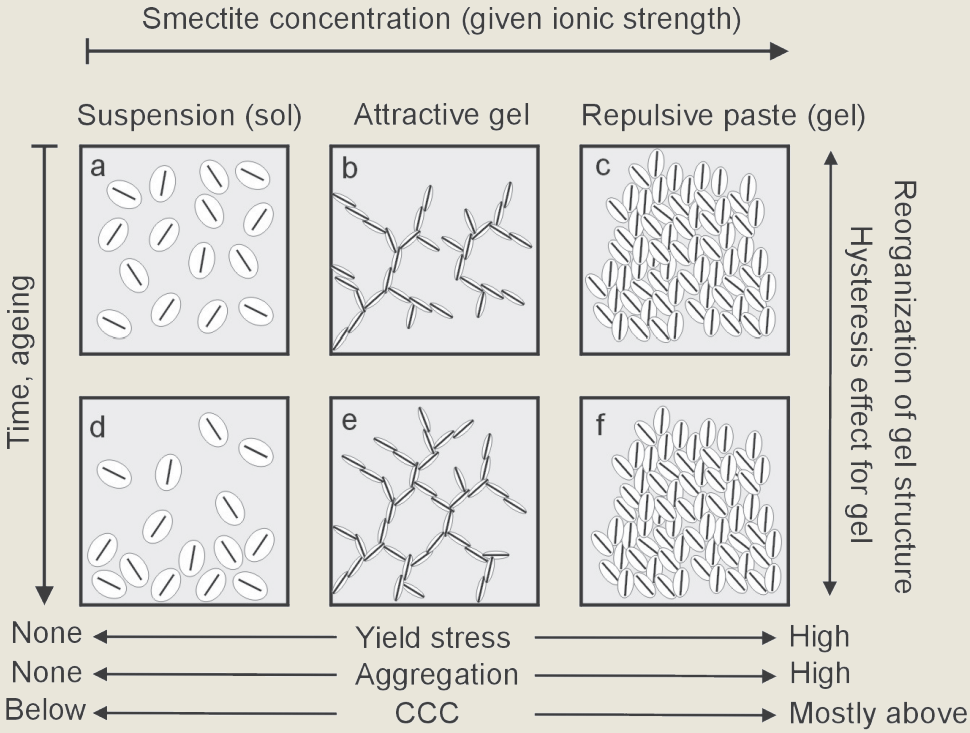
WP3: Radionuclide and host rock interactions



The objective of WP3 was to develop process level understanding of colloid mobility controlling processes and their appropriate description. This included radionuclide sorption and retention processes.
Figure 4 illustrates the processes considered in work package 3.

Figure 4 A schematic representation of the processes investigated within work package 3; namely (a) the radionuclide interaction with clay colloids and the observed metal-colloid dissociation rates (fast and slow dissociation step k3, k4) beside the direct radionuclide fracture surface interaction rate (k1) and the potential transfer rate from the fast dissociating to the slow dissociating mode as a function of metal-colloid contact time, (b) the mobility determining effect of fracture surface roughness and charge heterogeneity and (c) the effect of flow path complexity and matrix diffusion on colloid retention.

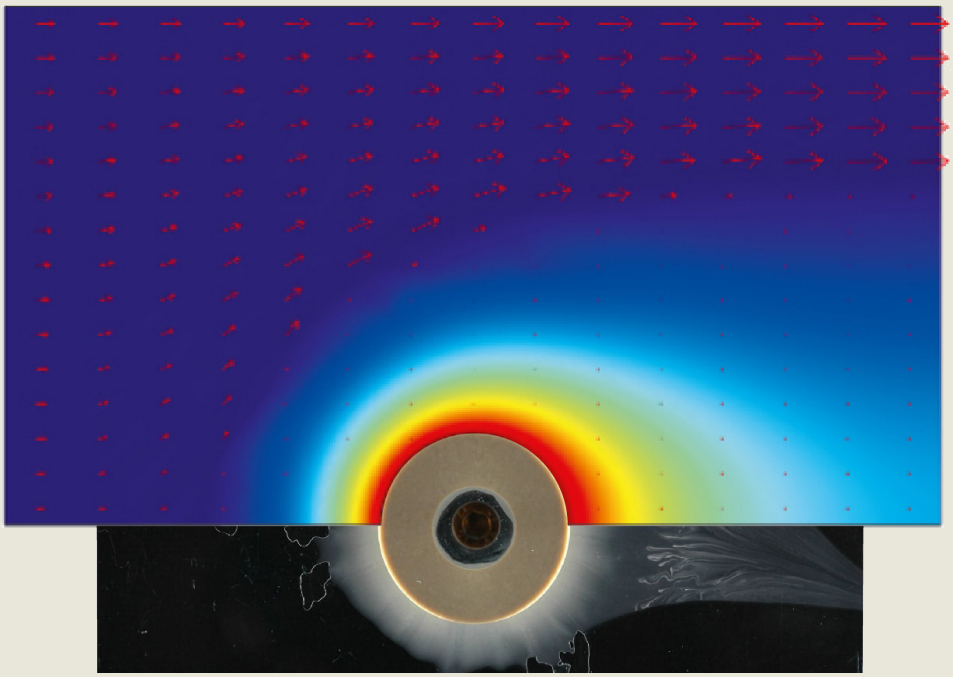
WP4: Colloid stability



The objective of WP4 was to understand clay colloid stability under different geochemical conditions with respect to ionic strength, pH and the influence of complexing agents. Figure 5 shows a summary representation of the underrating of the organization of smectite particles under different conditions.

Figure 5 *Schematic representation of Na-montmorillonite phases as function of montmorillonite concentration at given ionic strength investigated in WP4. a) montmorillonite particles are only weakly interacting, b) montmorillonite particles above CCC forms gel structure, c) at high montmorillonite concentration the repulsion forces dominate. Ageing d) sedimentation, e) and f) reorganization of gel structure."*

WP5: Conceptual and mathematical models



The objective of WP5 was 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 high-level radioactive waste and spent nuclear fuel.

Figure 6 *Examples of model results (top) and experimental observations of bentonite erosion.*

Outcome of BELBaR

The results from BELBaR shows that the current approach for treatment of colloid in most instances is reasonable and can be justified based on sound scientific understanding. There are some areas were the current approach is unnecessarily pessimistic (e.g. an assumption of de-ionized water), while there also are areas where there are uncaptured phenomena and additional research will be needed (e.g sedimentation in sloping fractures).

The following points represent a summary of recommendations[1]:

1. With respect to the safety assessment, the results of BELBaR suggest that assumptions made by WMOs related to the dependency of mass loss rate on groundwater flow velocity could potentially be reviewed under no highly 'erosive' conditions. It has been shown that bentonite erosion overall is driven more significantly by chemical forces rather than mechanical forces and thus it is recommended that the current treatment could be reviewed with respect to the driving mechanism controlling bentonite erosion.
2. All exchanger sites in the clay are currently assumed to be occupied by Na+. This assumption leads to high erosion rates and overly pessimistic mass loss calculations since the current modelling capability is not able to take into account the effects of divalent cations such as Ca2+. However, in a natural system, it would be difficult to exclude that the Na content of clay is less that the 20-25% threshold that favours colloid generation and thus the current treatment is considered to be appropriate within the overall system uncertainties.
3. The presence of different cations in solution can effect coagulation with divalent cations being more effective than monovalent ions as coagulants. It is currently assumed that mass loss ceases when groundwater salinity exceeds a stability limit of 4-8 mM NaCl for Na-bentonite. Should the assumed composition of the clay be reviewed (Recommendation 2) then consideration should also be given to addressing the conservatism that would be inherent in the assumed bentonite stability limit. However, within the bounds of the current knowledge, it considered that this assumption remains appropriate.
4. In terms of the ionic strength of groundwater assumed, given that deionised water may not be representative of a real dilute groundwater, it may be considered that the maximal zero charge limit is an overly conservative scenario. However it is considered that this assumption remains appropriate within the overall system uncertainties, particularly with regard to the need to address the potential for a change on groundwater composition for example due to glacial meltwater.
5. With respect to the angle of the fracture aperture, it is observed that the mass loss mechanisms between a horizontal and sloped fracture are different. Current performance assessment assumptions assume a horizontal fracture and thus the results observed suggest that this treatment may not necessarily be conservative. It is strongly suggested that this assumption is reviewed and potentially further work is required to account for the observed effects of slope angle/ gravity.
6. Significant retardation of clay colloids in a rock fracture has been observed. However, the uptake of colloids is not complete (i.e. some remain in solution), which means that colloid transport needs to be considered, if colloids are present. It is recommended that colloid retardation could be included in safety assessment calculations. However, the effect may be limited under most conditions and the process could therefore be optimistically neglected.
7. It is clear that the current assumption of linear sorption is valid. Sorption reversibility is nuclide specific and should possibly be treated on a case by case basis. However, for most radionuclides sorption reversibility can be assumed (the exception is potentially tetravalent elements where caution should be taken) over repository time scales.
8. Organic matter (humic or fulvic acid) was demonstrated to be able to stabilise clay colloids in NaCl electrolyte. In all the other electrolytes investigated (CaCl2, MgCl2) and at higher Ionic strength, the clay colloids undergo fast coagulation, independently of the presence of organic matter. This is true in various aqueous media containing different inorganic cations, showing that the ionic strength remains the key parameter. In addition, Ca2+ ions alone are able to initiate clay colloid agglomeration even at low concentrations. The presence of Ca, even at only low concentration in natural media, is thus recommended to be considered in performance assessments (as per Recommendations 2 and 3).
9. Regarding validation and advancing the models, sufficient confidence was obtained to predict clay mass loss rate in laboratory scales using numerical simulations whereas mass loss rate predictions in repository relevant scales remain to be assessed using analytically derived expressions for bounding estimates. According to the bounding estimates referred to, the agglomerate/floc migration rate in fractures is the mass loss rate determining feature.
10. The reasoning of dominant processes was succeeded considering agglomerate migration but was based only on expert judgement for clay swelling and gravity. Moreover, data needs specifications to assess the relative importance of clay swelling and gravity were raised but only when it was too late to commit experiments within BELBaR.
11. It can be stated that a new bounding estimate is proposed to be used in the safety cases to assess clay mass loss rates; loss of clay at the clay-water interface is limited by migration of newly formed clay agglomerates in fractures. This estimate can be obtained with far lesser efforts and uncertainties than used in the previous safety cases.

1 Recommendations may be considered most relevant to site specific assessments.

No.	Acronym	Name	Country
1	SKB	Svensk Kärnbränslehantering	SE
2	CIEMAT	Centro de Investigaciones Energeticas, Medioambientales y Technologicas	ES
3	NRI	Nuclear Research institute Rez plc	CZ
4	KIT	Karlsruhe Institut of Technology	GE
5	POSIVA	Posiva OY	FI
6	VTT	Technical Research Institute of Finland	FI
7	ClayTech	Clay Technology	SE
8	JYU	University of Jyväskylä	FI
9	KTH	Kungliga Tekniska Högskolan	SE
10	NDA/RWM	Nuclear Decommissioning Authority/ Radioactive Waste Management	GB
11	B+Tech	B+Tech	FI
12	UNIMAN	University of Manchester	GB
13	HU	Helsinki University	FI
14	MSU	Lomonosov Moscow State University	RU

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