

Bentonite analogue research related to geological disposal of radioactive waste: current status and future outlook

Heini M. Reijonen¹ · W. Russell Alexander²

Received: 20 September 2013 / Accepted: 13 March 2015 / Published online: 12 April 2015
© Swiss Geological Society 2015

Abstract The practice of utilising natural analogues in assessing the long-term behaviour of various components of geological repositories for radioactive waste is already well established in most disposal programmes. Numerous studies on bentonites, focussing on bentonite interaction with other components of the engineered barrier system and a range of host rock environments, are present in the literature. In this article, recent bentonite natural analogue studies are briefly reviewed, and gaps in the current literature identified, with the aim of (1) suggesting where relevant new information could be obtained by data mining published bentonite natural analogue studies with a new focus on current safety case requirements, (2) collecting relevant information by revisiting known bentonite analogue sites and conducting investigations with more appropriate analytical techniques, and (3) identifying novel study sites where, for example, bentonite longevity in very dilute to highly saline groundwater conditions can be studied. It must be noted that the use of natural analogues

in safety case development is likely to be site and repository design-specific in nature and thus emphasis is placed on the appropriate use of relevant natural analogue data on bentonite longevity.

Keywords Bentonite longevity · Smectite stability · Montmorillonite · Self-analogue · Examples from Finland, Sweden, Canada

1 Introduction

Bentonites are planned to be used in the engineered barrier systems (EBS) in most of the national high level waste (HLW) and spent nuclear fuel (SNF) disposal programmes (cf. Alexander et al. 2015, this issue), in crystalline, clay and evaporite host rock environments (cf. Alexander and McKinley 2007). The current status of those bentonite processes related to EBS interactions and bentonite buffer performance in various host rock environments has been recently reviewed in detail by Laine and Karttunen (2010) and Wilson et al. (2011). In this article, the previous overviews will be updated from the natural analogue (NA) viewpoint and broadened to cover other host rocks (such as clays and evaporites). Site-specific data on bentonite analogues related to crystalline rock environments and the KBS-3V¹ repository design have been recently reviewed as part of “complementary considerations” (see Reijonen et al. 2015, this issue) for repository performance included in the Olkiluoto (Finland) site safety case (Posiva 2012a). In general,

Editorial handling: A. G. Milnes.

This paper is published as part of the special theme: Natural Analogue Research for Deep Disposal of Nuclear Waste.

Most of the reports cited in this paper can be downloaded free of charge or ordered in hard copy from the website of the corresponding institution or company. The addresses can be found after the institution or company entry in Appendix 1 of the introductory review paper (Alexander et al. 2015, this issue).

✉ Heini M. Reijonen
heini.reijonen@sroy.fi

¹ Saanio & Riekkola Oy, Laulukuja 4, 00420 Helsinki, Finland

² Bedrock Geosciences, Veltheimerstrasse 18, 5105 Auenstein, Switzerland

¹ KBS-3V is the reference design for the repository planned to be constructed at Olkiluoto, Finland, in which the spent nuclear fuel canisters are emplaced in individual vertical deposition holes within a crystalline host rock.

the most relevant processes that can be evaluated via NA research related to bentonite EBSs are heat transfer, bentonite-groundwater interaction (salinity, pH, alkalinity), mechanical deformation and microbial activity. Additionally, processes that relate to interactions with other materials in the EBS (e.g. bentonite with the copper, steel or titanium containers, cementitious rock grouts etc.) are also of great importance. A glossary of terms and abbreviations used in this Special Issue is appended to the introductory review paper (Alexander et al. 2015, Appendix 1).

2 Definitions

2.1 Rock types and mineralogical composition

Bentonite is a common name for soft, plastic, porous, light-coloured rock composed mainly of clay minerals (from the smectite group) and colloidal silica produced by the chemical alteration of volcanic ash, either in situ or after transport (Hallsworth and Knox 1999; Fig. 1). Meunier (2005) defines bentonite as mono-mineralic clay rock, the smectite mineral being most commonly montmorillonite. Bentonites occur as a part of sedimentary strata of varying thickness and degree of diagenetic alteration and it is quarried worldwide for various industrial purposes due to its unique properties (Fig. 2).

From the HLW/SNF repository design point of view, the utilization of the term bentonite is more restricted than the geological term for bentonite sensu lato (see also discussion in Alexander et al. 2012). EBS designs often rely on very specifically defined bentonite compositions as the buffer material around the waste (e.g., in the Finnish case, the Na-montmorillonite content of the dry buffer material shall be 75–90 % by weight, Posiva 2012b). The range of the bentonite-based buffer and backfill materials considered covers also Ca-bentonites (e.g. SKB 2011) and clay-aggregate/sand mixtures (e.g., Man and Martino 2009).

Natural bentonites can thus differ significantly from the buffer bentonites intended for use in a geological repository. In addition to initial mineralogical composition, the production of the buffer material alters the original microstructure by grinding and re-compaction. The exchangeable cation composition of montmorillonite is also important regarding the buffer properties (i.e., related to swelling capability; e.g. Komine and Ogata 2003) and is known to vary even within individual bentonite deposits (e.g. Alexander and Milodowski 2014). Some industrial bentonites are also modified by changing the natural exchangeable cation composition. Natural bentonites also have accessory minerals in varying compositions depending on the deposit (cf. Wilson et al. 2011). In comparison, experimental work often relies on the use of purified

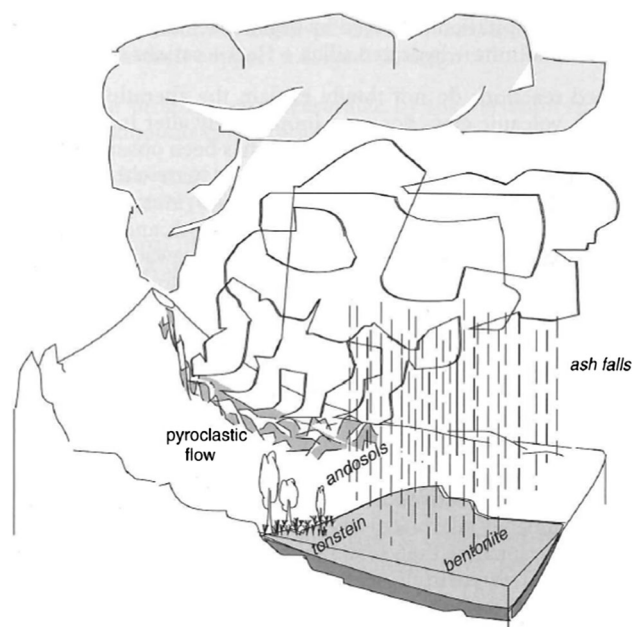


Fig. 1 Schematic presentation of alteration environments of ashes by Meunier 2005 (Fig. 7.10a). Bentonites form when volcanic ash is deposited in lakes, lagoons or shallow sea areas

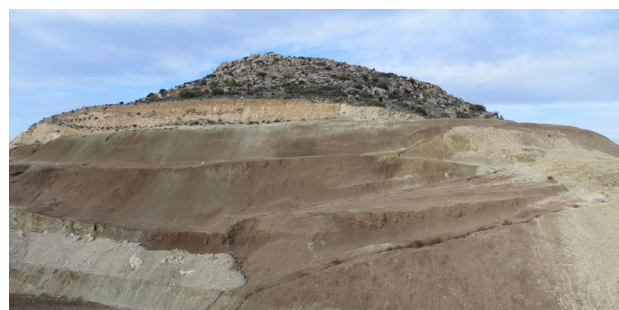


Fig. 2 Bentonite quarry near Kato Moni, Cyprus. Bentonite (greenish/reddish) is overlain by limestone cap rock and colluvium deposits

montmorillonite samples with fixed exchangeable cation composition. In addition to the mineralogical composition of bentonites and EBS components, density and inherent porosity are properties that have to be accounted for when assessing the behavior of these systems. All this leads to the conclusion that great care has to be taken when utilising bentonite data from the above mentioned disciplines (nature, laboratory) and in the use of terminology. Finally, as noted in Alexander and Milodowski (2014), the overall physical similarity of the dimensions of the natural analogue to the repository buffer bentonite is also crucial.

2.2 Features, events and processes (FEPs)

Relevant FEPs, to be taken into consideration when assessing long-term safety, are always case-specific and

have to be screened against the site information as well as the repository design (see e.g. Posiva 2012c). Common FEPs to all HLW/SNF repository designs which utilise bentonite buffer materials include heat generation from the SNF or HLW, groundwater-clay interaction and interactions between clay and other EBS materials, which may vary greatly depending on the concept and site adopted and the waste type to be disposed. In addition to FEP definitions, their couplings and interactions are of great importance in assessing long-term performance of repository systems.

3 State-of-the-art and future outlook

3.1 Introduction

In the space available, it is not possible to cover the recent activities in all disposal programmes worldwide so, here, two, quite different, examples are examined:

- The Finnish and Swedish disposal programmes where potential repository sites have been identified and where the EBS design is at a mature stage
- The UK programme, where both the site and the EBS design are at a purely generic stage and therefore the boundary conditions are less well defined

3.2 KBS-3 (Finnish and Swedish contexts)

In the case of the Finnish safety case, bentonite analogues serve as a source of information in the general process understanding (Posiva 2012c). The most significant sources of bentonite NA information have been screened in the Complementary Considerations (CC) report (Posiva 2012a; Reijonen et al. 2015, this issue) in order to enhance confidence in the safety case for the KBS-3V repository design. The thermal and hydrogeochemical constraints (and their couplings) on bentonite stability have also been recently reviewed by Laine and Karttunen (2010). Bentonite NAs were also utilized in Posiva's latest performance assessment (Posiva 2013) in relation to assessing the thermal stability of bentonite.

In comparison with the more technically focussed work of Laine and Karttunen (2010), the approach taken in CC report was to address the diverse and less quantifiable types of evidence and arguments in order to enhance confidence in the outcome of the safety assessment. In addition, the report attempted to demonstrate repository safety over the long-term, especially times greater than a few thousand years (Posiva 2012a; see also Reijonen et al. 2015, this issue). As such, a much wider view of NA studies was taken, with more use of qualitative examples to support

general design concepts and arguments about long-term safety. Overall, the conclusions noted that, while many examples of bentonite-chemical reactions exist, a more rigorous integration with laboratory data was required (see Sect. 3.4 and references therein and Posiva 2012a).

In the Swedish programme, bentonite analogues have been reviewed recently in relation to cement-bentonite interaction (Sidborn et al. 2014). Here, the focus was potential interaction between the EBS bentonites and leachates from cement grouts elsewhere in the repository and the approach taken was very much a modelling-based one, employing hydrogeological, hydrochemical and coupled (geochemical and transport) codes. As such, the bentonite NA input was used as a source of information to guide modelling efforts aimed at assessing the potential long-term impact on the repository performance (Sidborn et al. 2014). In addition, the NA data were used much in the form of a 'reality check', to establish the degree of confidence which could be placed in the modelling results by comparison with examples from relevant natural systems which have operated over repository-relevant timescales.

Generally, the study of reaction of bentonites in alkali solutions is a younger science than that of cement degradation and so, to some degree, the uncertainties are somewhat greater. When this is combined with the fact that experimental study of bentonite reaction is difficult due to a range of reasons, including the very fine grained nature of the primary bentonite minerals and the secondary reaction phases, and that even the definition of bentonite porewater chemistry is relatively novel, it is clear that the body of evidence available to constrain models of bentonite reaction remains highly limited (see discussion in Alexander and Milodowski 2014). In this paper, it was concluded that more information on the potential reaction of bentonite with alkali leachates is required.

3.3 UK context

The approach taken by Wilson et al. (2011) aims at providing tools that could be considered by the Nuclear Decommissioning Authority Radioactive Waste Management Directorate (NDA-RWMD), presently Radioactive Waste Management Limited (RWM) in an R&D (research and development) programme for buffer behaviour in relation to UK repository development and therefore covers a wider range of process consideration than the more site and design specific programmes in Finland and Sweden. The review focussed on the current understanding of the processes and on the possible requirements in design, experimental studies and modelling in any future UK programme. The most important conclusions in Wilson et al. (2011) are the fact that the processes described are likely to be highly coupled (cf. focus of Finnish

programme) and that there is a need for THMCB (Thermal/Hydraulic/Mechanical/Chemical/Biological) modelling ability.

3.4 Gaps identified and topics considered to be of interest for future research

Based on the literature reviews briefly discussed in the above sections and in a wider examination of NA requirements for the safety case (e.g. Posiva 2012a; Sidborn et al. 2014; Alexander et al. 2014, 2015, this issue), a comprehensive list of unresolved issues has been collated in the following Sects. 3.4.1–3.4.13. This section represents a brief overview of the main processes of relevance to the long-term behavior of bentonite in the repository EBS, backfill and seals and reported status of process understanding. In each section, first, the main bentonite processes of interest are listed and, in second, recommendations for addressing unresolved issues and open questions are identified.

3.4.1 Thermal effects

Process studied Thermal effects on bentonite have been studied through NAs, but insufficient information is available on the relevant temperature regimes (Posiva 2012a; Laine and Karttunen 2010) and this will need to be carefully considered in the design and development of the safety case (c.f. Wilson et al. 2011). This applies also to those repository designs where the canister surface temperature is >100 °C and thermal alteration could be an issue. Wyoming bentonites hold potential for both thermal and hydrogeochemical studies (Laine and Karttunen 2010).

Future outlook Return to more relevant sites (e.g., Busachi in Sardinia; cf. Laine and Karttunen 2010) and apply a range of more relevant analytical techniques to better understand the site history and temperature profiles. It is proposed here that one or two more appropriate sites be revisited and a range of more relevant analytical techniques be applied to better understand the site history and deposit temperature profiles. Only then should the site be re-sampled and the standard clay analyses previously carried out repeated. In this manner, the resultant data can be placed in much better context and any extension to the repository environments be better grounded. It is acknowledged that, in particular, the availability of potassium should be better constrained in order to estimate controls over smectite/illite reactions in the repository environment. In addition, the Clay Spur bed (Wyoming) should be studied in order to better constrain the thermal and hydrogeological boundary conditions preserving the deposit.

3.4.2 Bentonite resaturation

Process studied Large-scale experiments suggest that theoretical and laboratory-based understanding of bentonite resaturation may not be an effective description of the process at repository scale. Resaturation times may be significantly longer than previously predicted and full resaturation may not occur at all in some cases (c.f. Wilson et al. 2011; Alexander and Milodowski 2014).

Future outlook Boundary conditions for dry and wet thermal reactions can be obtained via new, better constrained, NA studies at existing sites (such as Busachi in Sardinia).

3.4.3 Bentonite plasticity

Process studied Current evidence of bentonite plasticity is of value, but the topic has not been treated quantitatively enough to warrant use in a safety assessment (Posiva 2012a).

Future outlook The approach of Keto (1999) should be repeated plus the process should be included in current URL experiments (e.g. FEBEX; Gens et al. 2002). In addition, an attempt should be made to find a site where canister sinking could be studied by means of boulder fall/overburden pressure on bentonite.

3.4.4 Iron-bentonite interaction

Process studied In the case of iron-bentonite interactions, useful NA data are very limited (cf. Marcos 2003; Wersin et al. 2007). Steel/iron corrosion products could react with bentonite to induce reduced swelling of the montmorillonite or produce non-swelling minerals, thereby causing changes in swelling pressure, bentonite cementation and loss of radionuclide sorption sites. The kinetics of smectite alteration to iron-rich minerals is not well understood (Wilson et al. 2011).

Future outlook Existing NA studies are not relevant enough for existing repository designs, although general observations from natural systems and the NF-PRO URL experiment (Kickmaier et al. 2005) suggest uptake of iron could reduce swelling pressures. These data need to be integrated with a more appropriate NA study. There is identified need for future studies (such as presented by Pelayo et al. 2011).

3.4.5 Copper-bentonite interaction

Process studied Cu-bentonite interaction: only limited information available (Posiva 2012a).

Future outlook From Kronan cannon, qualitative data are available and these should be more rigorously integrated with existing/new laboratory data.

3.4.6 Glass-bentonite interaction

Process studied Effect on bentonite of waste glass (vitrified HLW) in designs with no container, or very thin container, is not understood. Glass dissolution may increase in contact with some clays, but mechanisms are poorly understood (Wilson et al. 2011).

Future outlook Sites where natural glasses are in contact with bentonites exist. However, the differences between natural glasses and vitrified radioactive waste must not be overlooked (cf. Miller et al. 2000).

3.4.7 Freezing/thawing processes

Process studied On the bentonite buffer freezing and thawing processes, only general considerations are available (Posiva 2012a).

Future outlook A NA study of bentonite in a glaciated terrain could provide more reliable tools for extrapolating the existing experimental evidence to safety case time scales.

3.4.8 Chemical erosion

Process studied There is no NA study to support or discount the process (dilute conditions during glacial melt water intrusion; Posiva 2012a). Further research and development work is likely to be needed on erosion of bentonite, especially for specific conditions (Wilson et al. 2011).

Future outlook Montmorillonite has been observed to occur as a fracture filling mineral (e.g. at repository sites such as Olkiluoto in Finland and Forsmark in Sweden), providing potential in situ NA sites for montmorillonite stability under varying hydrogeochemical conditions (see discussion in Sect. 4) and many other sites for similar studies have been identified. Additionally, studies related to the erosional histories of known bentonite deposits under the effects of the last glaciation or by meteoric waters would be of interest.

3.4.9 Saline groundwater

Process studied Saline groundwater conditions may need to be further considered (Wilson et al. 2011). The impact of temperature gradients on the accumulation and dissolution of salts in the vicinity of canisters embedded in clay buffer also needs to be assessed. Experimental/modelling/NA work may be required to increase understanding

if bentonite is to be exposed to unusually high Mg concentrations (i.e. Mg-rich brine).

Future outlook Approach proposed in Sect. 4 is worthwhile for bentonite-saline groundwater interaction and understanding the impact of Mg-rich brines. Temperature gradient impact on salt accumulation could be addressed at Busachi (see Sect. 3.4.1).

3.4.10 Cement-bentonite interaction (including low alkali cement)

Process studied OPC cement-bentonite interaction: experimental work to reduce uncertainties concerning smectite alteration rates and bentonite degradation rates under highly alkaline conditions needs to be extended to include integrated NA studies (Sidborn et al. 2014). With respect to low alkali cement-bentonite interaction, NA studies in Cyprus have been finalised recently (Alexander and Milodowski 2014) and are currently ongoing in the Philippines (Alexander et al. 2008).

Future outlook There is little hard evidence on OPC-bentonite interaction from NAs, but the topic could be addressed at the Khushaym Matruk site in Jordan (see discussion in Pitty and Alexander 2011). In addition, in Cyprus, there is a site where pH 12 ground waters in contact with clays have been observed (see Alexander et al. 2011 for details). Results from Cyprus clearly show limited reaction for low alkali cement-bentonite interaction; additional information from the ongoing Philippines study can contribute to confidence on the process understanding.

3.4.11 Coupled processes (including THMBC)

Process studied A key point to note is that, although for simplicity the reactions and processes are discussed as individual topics, many of the processes of relevance to the long-term behaviour of bentonite will be strongly coupled and cannot be considered, or optimised, in isolation. Despite recent advances through the EU projects DECOVALEX, FORGE (see glossary in Alexander et al. 2015, this issue) and PEBS project (Schäfers and Fahland 2014), there remains a general need to increase understanding of coupled processes, as has been noted in the latest stage of DECOVALEX. Assessment of clay materials with respect to coupled THMBC behaviour (including gas conductivity and ion diffusivity) needs to be made in order to obtain a basis for optimal selection of one specific candidate buffer/backfill material (Wilson et al. 2011).

Future outlook Focussing on coupled processes should be introduced more strongly in NA research in general. Potential sites exist e.g. for cement-bentonite alteration coupled with thermal alteration and these should be studied.

3.4.12 Bentonite–sand/rock mixtures

Process studied Performance of bentonite–sand/rock mixtures are being considered more as repository designs are optimised.

Future outlook In reality, the physical and mineralogical properties of many natural bentonites are more akin to bentonite–sand or bentonite–rock mixtures (see discussion in Alexander and Milodowski 2014), allowing the long-term behaviour of these mixtures to be assessed quantitatively. To date, only a tentative assessment has been carried out, but this has shown the potential for further, detailed studies.

3.4.13 Smectites other than montmorillonite

Process studied Consideration of smectites other than montmorillonites: when assessing other clays (e.g., saponite, Wilson et al. 2011 or Friedland clay, Posiva 2012b), the first action should be a study of the source deposit and what information can be gleaned on the long-term behaviour of the clay under repository-relevant conditions (c.f. Clay Spur studies for MX-80 bentonite; Laine and Karttunen 2010).

Future outlook Alteration patterns for natural trioctahedral smectites such as saponite are well known, providing significant potential for NA studies of alternative buffers under a range of geological environments.

3.4.14 Microbial processes

Process studied It has been generally accepted that the bentonite buffer would minimise microbial populations due to the restriction of nutrient supply via groundwater flow, but more recent studies (e.g. Stroes-Gascoyne 2010) of several bentonite-backfill and sealing materials used in large-scale experiments at AECL's Whiteshell URL, have demonstrated the presence of viable microbial populations in all bulk materials.

Future outlook These few, URL-based examples should be verified by examining microbial populations in natural bentonites. This could most easily be carried out in conjunction with studies on other aspects of bentonite (e.g. saline reaction). Certainly there is a suggestion of microbial processes in the observed bentonite reactions studied in the Philippines (Arcilla et al. 2011).

4 Stability of smectites in crystalline and sedimentary environments: the self-analogue concept

In many cases identified in Sect. 3, there is clearly the potential to increase process understanding via NA studies. As an example of the processes addressed in Sect. 3.4, one particular and novel approach using smectite stability in fresh to saline ground waters as a “self-analogue” is addressed a little more detail below.

Bentonite buffer stability has to be assessed, depending on the site, for various groundwater conditions from dilute (especially in relation to glacial meltwater intrusion and the process of chemical erosion of bentonite) to saline, and even brine conditions, the former being the expected range at most crystalline sites and the latter more relevant to deep sedimentary and evaporite sites. Hence, in addition to NA studies of bentonite deposits, smectites and their longevity at different sites which are similar to a repository site and under different groundwater conditions can be investigated as what is here referred to as “self-analogues”.

The potential for using natural host rock smectites to assess the likely longevity of industrial EBS bentonite was initially raised by Arthur and Savage (2012) (see also Savage 2012) when reviewing information on the Forsmark site for the Swedish regulator (SSM). They noted that smectite and calcite occur at all depths in Forsmark fractures (see Fig. 3), with no evidence for removal/dissolution by previous glacial episodes. Smectites have been reported from candidate repository sites in Finland, Sweden and Canada (e.g. Gehör 2007; Sandström et al. 2008; Koroleva et al. 2009 respectively). In Finland, montmorillonite has been reported from the fracture gouges sampled at the underground rock characterisation facility ONKALO. Occurrences have also been reported at numerous other Finnish sites (e.g. Front et al. 1998), suggesting smectite may be widespread in other geological environments similar to the Olkiluoto host rock.

Dating of the fracture systems suggests that the smectites have been present since at least the Palaeozoic (i.e. 540–250 Ma BP) with the deep brines in contact with the smectites having been stable for at least several million years, if not longer (Smellie et al. 2008). The fact that the smectite exists in these formations is already a powerful argument for the longevity of bentonite, although this point has not been discussed in depth to date. To provide a high degree of confidence in such an argument, it requires that it be fully backed by scientific evidence and so needs to be studied in detail in future.

Smectite occurrences have also been reported for samples from the Oskarshamn site in Sweden (Drake et al. 2006) and montmorillonite at all depths in Hästholmen (Gehör et al. 1997a; Front et al. 1999), Kivetty (Gehör et al.

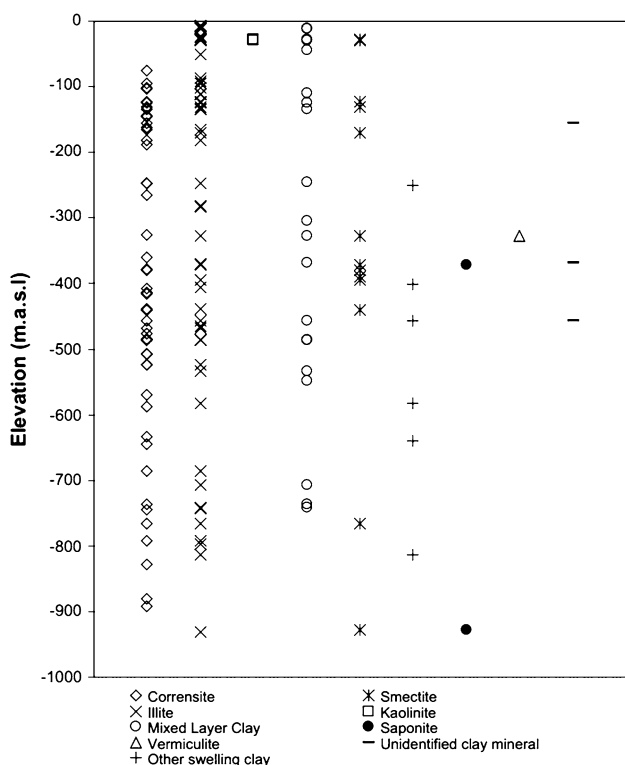


Fig. 3 Depth distribution of different clay minerals identified by X-ray diffraction studies carried out at the Forsmark site, Sweden (Sandström et al. 2008)

1997b) and Romuvaara (Kärki et al. 1997) study sites in Finland, showing the widespread abundance of smectite minerals in the Fennoscandian Shield. The abovementioned locations vary from inland (e.g. Romuvaara) to coastal (e.g. Olkiluoto and Forsmark). Deep groundwater systems in the Fennoscandian bedrock have been studied in great detail in relation to repository site investigations and these data provide the salinity range for conditions where smectites occur (Fig. 4). In addition, the spatial and temporal evolution of the groundwater salinities has been studied in detail (Fig. 5), so an assessment of likely previous groundwater salinities at any given borehole depth can be conducted. This will allow the impact of a wide range of salinities on the smectite longevity to be elucidated when the mode of occurrence of smectite in relation to specific groundwater systems has been defined.

In Canada, at the Bruce site in the Michigan Basin of North America, (Fig. 6) mixed layer illite/smectites have been reported (e.g. Koroleva et al. 2009) in the Ordovician Shales above the potential repository host formation. It would appear that the lower members of the shale (Blue Mountain and Collingwood) have gone through the oil window (e.g., Béland Otis 2012) and this is assumed to have converted any smectite originally present to illite (Jackson 2009). However, mixed layer illite/smectite has been

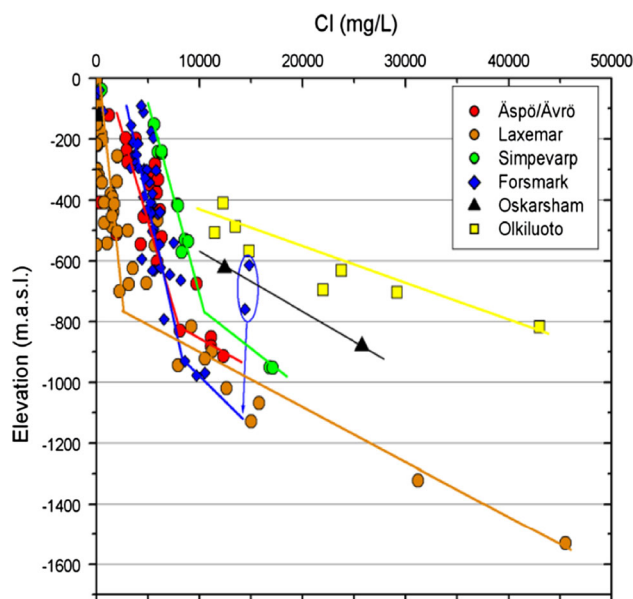


Fig. 4 Groundwater chlorinities at various sites in Sweden and Finland as a function of depth (Smellie et al. 2008)

reported in the stratigraphically higher members (Queenston, Georgian Bay) where the groundwater/porewater total dissolved solids (TDS) reach as high as 300 gL^{-1} , somewhat higher than in the repository horizon (cf. Raven et al. 2010).

Recent work (e.g. Clark et al. 2013) on the palaeohydrogeology of the Michigan Basin suggests that the brines in the Bruce repository host formation are Palaeozoic in age, indicating long-term isolation of this groundwater system. It would therefore be useful to define precisely why the smectites are absent from the Collingwood horizon, while having been preserved in the Queenston and Georgian Bay horizons, as this could support the posit of longevity of bentonite under such highly saline conditions.

Fracture gouge minerals and smectites present in sedimentary rocks therefore provide the potential to study a self-analogue for the bentonite buffer considering a large range of hydrogeochemical conditions. Rigorous mapping and analysis of the detailed smectite mineralogy and properties, mineral paragenesis and relationship with groundwater (geochemistry and hydrogeology) can potentially provide a robust basis for qualitative support for the long-term buffer performance at the selected sites. In addition, it may be possible to quantify specific ranges for smectite stability fields over geological timescales.

5 Conclusion

This brief overview of data from several recent NA studies on natural occurrences of bentonite has shown that, although most processes relevant to the long-term

Fig. 5 Sketch showing tentative salinities and groundwater-type distributions versus depth for the transmissive zones at the Forsmark site, Sweden (Smellie et al. 2008). From left to right: **a** situation prior to the last deglaciation, **b** intrusion of the last deglaciation meltwater, **c** Littorina Sea water penetration caused by density intrusion (or turnover), and **d** present day situation

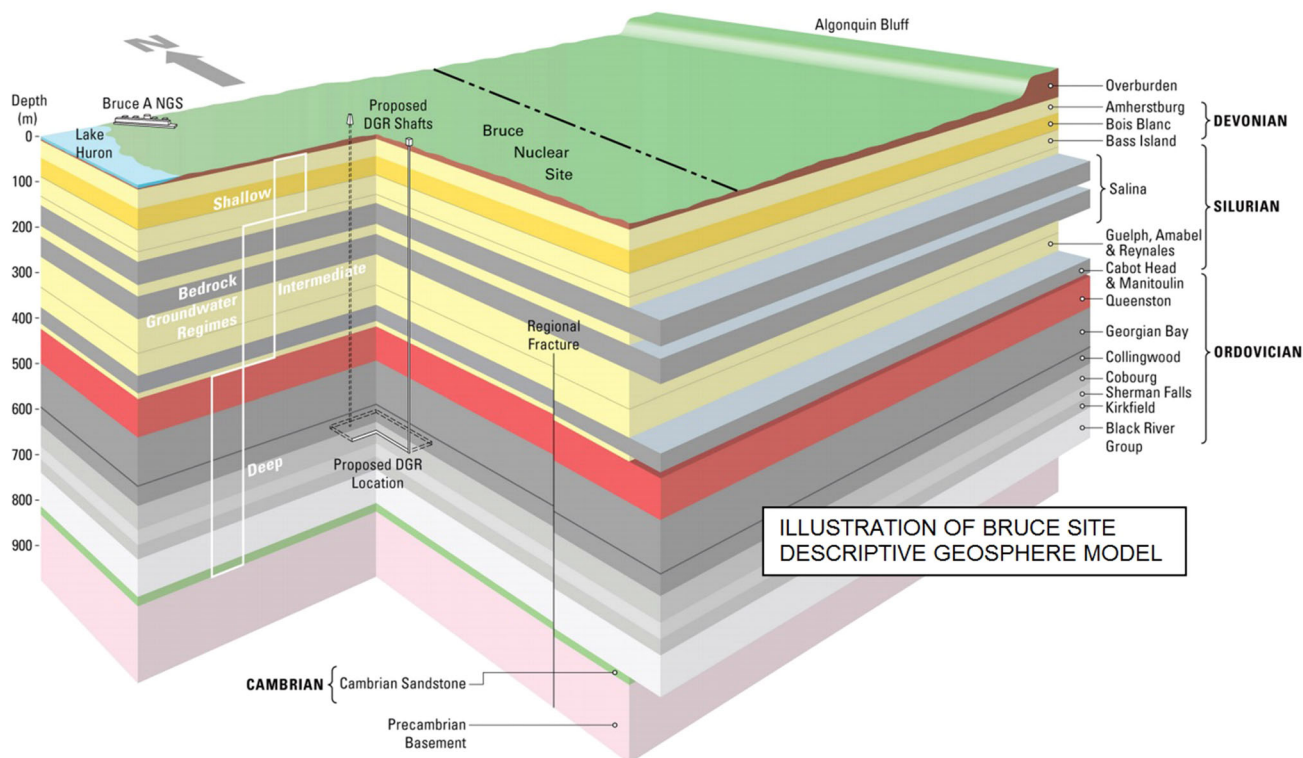
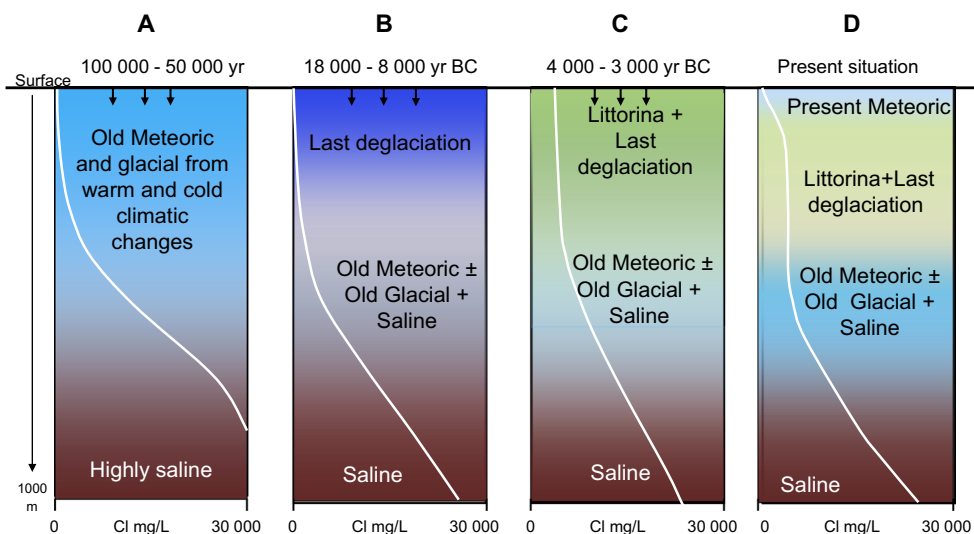


Fig. 6 Descriptive geosphere model of the Bruce site (Jensen et al. 2009)

behaviour of EBS bentonite under repository conditions could be addressed by NA studies (Sect. 3), few have been carried out to an appropriate level to date. Either the boundary conditions have not been fully defined, or the environments studied were not as relevant as hoped, or the studies were simply not focussed enough on supplying data for the safety case (cf. Alexander et al. 2014). This is reflected in the strength of the qualitative discussion and

strong process understanding regarding engineered clay barriers and their longevity in general but, on the other hand, in the lack of usable quantitative data for safety assessment models.

Nevertheless, many bentonite NAs of relevance do exist and much information of use in the safety case, both quantitative and qualitative, could be provided. These data could be produced by a mixture of means, including:

- Obtaining relevant information by data mining published NA studies with a new focus on current safety case requirements
- Obtaining relevant information by revisiting known bentonite analogue sites and conducting investigations with more appropriate analytical techniques
- Identifying novel study sites where, for example, long-term stability of bentonite in very low salinity ground waters can be studied

It is clear from the comparison of the Fennoscandian programmes and work contributory to the UK programme that the precise use of NA data is partly dependent on the maturity of the programme, but overall the NA approach has been shown to be flexible enough to address both ends of the spectrum of activities examined here.

Acknowledgments The authors would like to thank the reviewers, S. Norris and W. Steinger, for their thoughtful input.

References

- Alexander, W.R., Arcilla, C.A., McKinley, I.G., Kawamura, H., Takahashi, Y., Aoki, K., Miyoshi, S. (2008). A new natural analogue study of the interaction of low-alkali cement leachates and the bentonite buffer. *Scientific Basis of Nuclear Waste Management XXXI*, 493–500.
- Alexander, W. R., & McKinley, L. E. (Eds.). (2007). *Deep geological disposal of radioactive wastes*. Amsterdam: Elsevier.
- Alexander, W.R., McKinley, I.G., Kawamura, H. (2014). The process of defining an optimal natural analogue programme to support national disposal programmes. *Proceedings NEA-GRS Workshop on natural analogues for Safety Cases of repositories in rock salt. 4–6 September, 2012, Braunschweig, Germany. Radioactive Waste Management NEA/RWM/R(2013)*, (pp. 29–43). Paris: NEA/OECD.
- Alexander, W.R., Milodowski, A.E. (eds.) (2014). Cyprus natural analogue project (CNAP) phase IV final report. *Posiva Working Report 2014-02*. Eurajoki: Posiva.
- Alexander, W.R., Milodowski, A.E., Pitty, A.F. (eds.) (2012). Cyprus natural analogue project (CNAP) phase III final report. *Posiva Working Report 2011-77*. Eurajoki: Posiva.
- Alexander, W.R., Reijonen, H.M., McKinley, I.G. (2015). Natural analogues: studies of geological processes relevant to radioactive waste disposal in deep geological repositories. *Swiss Journal of Geosciences*, 108. doi:10.1007/s00015-015-0187-y.
- Arcilla, C. A., Pascua, C. S., & Alexander, W. R. (2011). Hyperalkaline springs in the Philippines: significance to natural Carbon Capture and Sequestration. *Energy Procedia*, 4, 5093–5100.
- Arthur, R., Savage, D. (2012). Equilibrium constraints on buffer erosion based on the chemistry and chemical evolution of glacial meltwaters. *Proc. of 5th International meeting on Clays in Natural and Engineered Barriers for Radioactive Waste Confinement*. 22-25 October, 2012, Montpellier.
- Béland O.C. (2012). Preliminary results: potential ordovician shale gas units in southern Ontario. Summary of field work and other activities 2012. In: Ontario Geological Survey, *Open File Reports 6280* (pp. 29-1–29-12). Ottawa: Ontario Geological Survey, Geology Ontario.
- Clark, I.D., Al, T., Jensen, M., Kennell, L., Mazurek, M., Mohapatra, R., Raven, K.G. (2013). *Paleozoic-aged brine and authigenic helium preserved in an ordovician shale aquiclude*. *Geology*. Boulder: Geological Society of America. doi:10.1130/G34372.1.
- Drake, H., Sandström, B., Tullborg, E.-L. (2006). *Mineralogy and geochemistry of rocks and fracture fillings from Forsmark and Oskarshamn: compilation of data for SR-Can. SKB R-06-109*. Stockholm: Swedish Nuclear Fuel and Waste Management Co. (SKB).
- Front, K., Paulamäki, S., Ahokas, H., Anttila, P. (1999). Lithological and structural bedrock model of the Hästholmen study site, Loviisa, SE Finland. *POSIVA Report 99-31*. Helsinki: Posiva Oy.
- Front, K., Paulamäki, S., Paananen, M. (1998). Updated lithological bedrock model of the Olkiluoto study site, Eurajoki, southwestern Finland. *Posiva Working Report 98-57*. Helsinki: Posiva Oy. (in Finnish with English abstract).
- Gehör, S. (2007). Mineralogical characterization of gouge fillings in ONKALO facility at Olkiluoto. *Posiva Working Report 2007-33*. Olkiluoto: Posiva Oy.
- Gehör, S., Kärki, A., Suoperä, S., Taikina-aho, O. (1997b). Kivetty, Äänekoski: petrology and low temperature minerals in the K1-K11 drill core samples. *Posiva Working Report 97-16*. Helsinki: Posiva Oy.
- Gehör, S., Kärki, A., Taikina-aho, O. (1997a). Loviisa, Hästholmen: petrology and low temperature fracture minerals in drill core samples HH-KR1, HH-KR2 and HH-KR3. *Posiva Working Report 97-40*. Helsinki: Posiva Oy.
- Gens, A., Guimaraes, L. D. N., Garcia-Molina, A., & Alonso, E. E. (2002). Factors controlling rock-clay buffer interaction in a radioactive waste repository. *Engineering Geology*, 64, 297–308.
- Hallsworth, C.R., Knox, R.W.O'B. (1999). BGS rock classification scheme. Volume 3: classification of sediments and sedimentary rocks. *British Geological Survey Research Report, RR99-03*. Nottingham: British Geological Survey, Keyworth.
- Jackson, R. (2009). Organic geochemistry and clay mineralogy of DGR-3 and DGR-4 core. DGR site characterization document Intera Engineering project 08-200. *Intera TR-08-29*. Ottawa: Intera.
- Jensen, M., Lam, T., Luhowy, D., McLay, J., Semec, B., & Frizzell, R. (2009). Overview of Ontario power generation's proposed L&ILW deep geologic repository Bruce site, Tiverton, Ontario. In M. Diederichs & G. Grasselli (Eds.), *ROCKENG09: Proceedings of the 3rd CANUS Rock Mechanics Symposium, Toronto, May 2009*. Toronto: Canadian Rock Mechanics Association.
- Kärki, A., Gehör, S., Suoperä, S., Taikina-aho, O. (1997). Romuvaara, Kuhmo: petrology and low temperature fracture minerals in the R0-R11 drill core samples. *Posiva Working Report 97-19*. Helsinki: Posiva Oy. (In Finnish with English abstract).
- Kickmaier, W., Vomvoris, S., & McKinley, I. G. (2005). Radwaste management: brothers Grimsel. *Nuclear Engineering International*, 50(607), 10–13.
- Komine, H., & Ogata, N. (2003). New equations for swelling characteristics of bentonite-based buffer materials. *Canadian Geotechnical Journal*, 40, 460–475.
- Koroleva, M., de Haller, A., Mäder, U., Waber, H.N., Mazurek, M. (2009). Borehole DGR-2: pore-water investigations. *Technical Report TR-08-02*. Switzerland: Rock-Water Interaction, Institute of Geological Sciences, University of Berne.
- Laine, H.M., Karttunen, P. (2010). Long-Term stability of Bentonite: a literature review. *Posiva Working Report 2010-53*. Eurajoki: Posiva.
- Man, A., Martino, J.B. (2009). *Thermal, hydraulic and mechanical properties of sealing materials*. NWMO TR-2009-20. Toronto: Nuclear Waste Management Organisation (NWMO).
- Meunier, A. 2005. *Clays* (pp. 472). Berlin Heidenberg: Springer-Verlag.

- Miller, W.M., Alexander, W.R., Chapman, N.A., McKinley, I.G., Smellie, J.A.T. (2000). *Geological disposal of radioactive wastes and natural analogues*. Waste management series, 2. Amsterdam: Pergamon.
- Pelayo, M., García-Romero, E., Labajo, M. A., & Pérez del Villar, L. (2011). Occurrence of Fe–Mg-rich smectites and corrensite in the Morron de Mateo bentonite deposit (Cabo de Gata region, Spain): a natural analogue of the bentonite barrier in a radwaste repository. *Applied Geochemistry*, 26, 1153–1168.
- Pitty, A.F., Alexander, W.R. (eds.) (2011). A natural analogue study of cement buffered, hyperalkaline groundwaters and their interaction with a repository host rock IV: an examination of the Khushaym Matruk (central Jordan) and Maqarin (northern Jordan) sites. *Bedrock Geosciences Technical Report 11-02*. Harwell: NDA-RWMD.
- Posiva. (2012a). Safety case for the disposal of spent nuclear fuel at Olkiluoto—Complementary Considerations. *POSIVA Report 2012-11*. Eurajoki: Posiva.
- Posiva. (2012b). Safety case for the disposal of spent nuclear fuel at Olkiluoto—Description of disposal system. *Posiva report 2012-05*. Eurajoki: Posiva.
- Posiva. (2012c). Safety case for the disposal of spent nuclear fuel at Olkiluoto—features, events and processes. *POSIVA report 2012-07*. Eurajoki: Posiva.
- Posiva. (2013). Safety case for the disposal of spent nuclear fuel at Olkiluoto—performance assessment. *POSIVA Report 2012-04*. Eurajoki: Posiva.
- Raven, K.G., Sterling, S.N., Jackson, R.E., Avis, J.D., Clark, I.D. (2010). Geoscientific site characterization of the proposed Deep Geologic Repository, Tiverton, Ontario. *Proceedings of Geo Canada 2010 Convention “Working with the Earth—Terre d’Avenir”*, Calgary, May 10 to 14, 2010. Canada: University of Calgary.
- Reijonen, H.M., Alexander, W.R., Marcos, N., Lehtinen, A. (2015). Complementary considerations in the safety case for the deep repository at Olkiluoto, Finland: support from natural analogues. *Swiss Journal of Geosciences*, 108. doi:10.1007/s00015-015-0181-4.
- Sandström, B., Tullborg, E.-L., Smellie, J.A.T., MacKenzie, A.B., Suksi, J. (2008). *Fracture mineralogy of the Forsmark site: Final report. SKB R-08-102*. Stockholm: Swedish Nuclear Fuel and Waste Management Co. (SKB).
- Savage, D. (2012). Geochemical Constraints on Buffer Pore Water Evolution and Implications for Erosion. *SSM report 2012:61*. Sweden: Swedish Radiation Safety Authority (SSM).
- Schäfers, A., Fahland, S. (eds.) (2014). *International Conference on the Performance of Engineered Barriers*. 6-7 February, 2014, EU PEBS Project. Hannover: Bundesanstalt für Geowissenschaften und Rohstoffe (BGR).
- Sidborn, M., Marsic, N., Crawford, J., Joyce, S., Hartley, L., Idiart, A., de Vries, L.M., Maia, F., Molinero, J., Svensson, U., Vidstrand, P., Alexander, W.R. (2014). *Potential alkaline conditions for deposition holes of a repository in Forsmark as a consequence of OPC grouting. SKB R-12-17*. Stockholm: Swedish Nuclear Fuel and Waste Management Co. (SKB).
- SKB (2011). Long-term safety for the final repository for spent nuclear fuel at Forsmark. *Main report of the SR-site project. SKB TR-11-01*. Stockholm: Swedish Nuclear Fuel and Waste Management Co. (SKB).
- Smellie, J.A.T., Tullborg, E.-L., Nilsson, A.-C., Sandström, B., Waber, N.H., Gimeno, M., Gascoyne, M. (2008). *Explorative analysis and expert judgement of major components and isotopes. SKB R-08-84*. Stockholm: Swedish Nuclear Fuel and Waste Management Co. (SKB).
- Stroes-Gascoyne, S. (2010). Microbial occurrence in bentonite-based buffer, backfill and sealing materials from large-scale experiments at AECL’s Underground Research Laboratory. *Applied Clay Science*, 47, 36–42.
- Wersin, P., Birgersson, M., Olsson, S., Karnland, O. & Snellman, M. (2007). Impact of corrosion-derived iron on the bentonite buffer within the KBS-3H concept. The Olkiluoto case study. *POSIVA Report 2007-11*. Olkiluoto: Posiva Oy.
- Wilson, J., Savage, D., Bond, A., Watson, S., Pusch, R., Bennett, D. (2011). Bentonite: a review of key properties, processes and issues for consideration in the UK context. *Quintessa Report QRS-1378ZG-1.1*. Henley-on-Thames: Quintessa.