6. Thermal alteration of bentonite (TAB): isn't it time we got it right?

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Introduction

In the case of thermal alteration of bentonite, cementation, caused by smectite/illite conversion where silica is released at higher temperature (and later reprecipitated) can occur. Currently, in many national programmes, the requirements for maximum bentonite temperatures are set around 100 °C (e.g. Posiva 2012a). In many cases, there would be interest in increasing this limit, so allowing, for example, the placement of waste packages more closely than is currently planned. But this would require more stringent justification than currently exists.

Although Miller et al. (2000, 2006) and Laine & Karttunen (2010) noted a significant number of NA papers on this theme in the literature, a recent re-analysis (Posiva, 2013b) shows that most examples suffer from the same problem: the conditions are not truly representative of a repository. In most cases of contact metamorphism studied, the temperatures have been much too high (800-900°C rather than the repository relevant maximum of 100°C in the current Finnish design; Posiva, 2013a). As illustrated in Figure 1, the degree of saturation of the EBS affects the thermal conductivity of the bentonite, the highest temperatures being met in the "dry" case, i.e. bentonite water contents being as installed. In Posiva's reference design, the initial water content of the buffer is assumed to be 17% (Posiva 2012a).

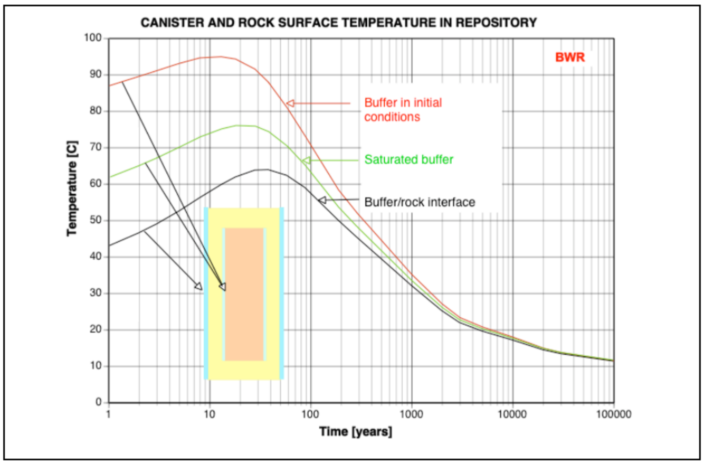

Figure 1: Likely temperature evolution in the bentonite buffer at Olkliluoto (for BWR fuel in a centrally located deposition hole; Ikonen & Raiko 2012).

Thermal-induced cementation: some examples

Several NA studies (e.g. Pellegrini et al., 1999; Woods et al., 2000) have tried to address the potential impact of thermally-induced cementation. Usually, smectite-bearing clays which had been penetrated by dykes/sills were examined and, at distance from the dyke where the temperature gradients were repository relevant, mineralogical changes were associated with subsequent fracturing of the cemented clays as the interstitial waters were driven out. Retrograde or 'back' reactions during the temperature decrease following the thermal event was suggested as the reason why new smectite was produced.

Unfortunately, these observations cannot directly be applied to the safety case because the full thermal, chemical and pressure histories experienced by these clays were not adequately characterised.

Pusch & Karnland (1988) investigated bentonite from the Busachi site in Sardinia and ran a series of calculations to assess the likely temperature at different places in the bentonite. They provided evidence that significant heat-induced dissolution of smectite occurred at 150 to 200ºC (i.e. much warmer than is likely in the buffer at Olkiluoto) and precipitation of siliceous material occurred during cooling. This siliceous cementation was found to have measurably affected the rheological properties of the bentonite, in a manner which might adversely affect the containment of radionuclides if it occurred in a repository environment.

However, the calculations were based on a range of assumptions about the site and the bentonite and these need to be ground-truthed before the results can be used quantitatively in a safety case.

Kolaříková & Hanus (2008) reported on the Ishrini bentonite body in Libya where the impact of a basalt dome and dykes on the bentonite was assessed by examining isotopic and mineralogical changes. It would appear that the minimum temperature experienced by the bentonite was probably >190°C but bentonite from outwith this zone (i.e. in the lower temperature area) was not sampled (Laine & Karttunen, 2010).

In conclusion, while the probable impact of raised temperatures is minimal (i.e. some local cementation occurs, but the majority of the bentonite remains unaltered), a truly Olkiluoto-representative analogy has not yet been examined.

Thermal-induced illitisation: some examples

In the case of diagenetic illitisation, a number of natural analogue studies have been carried out in the Gulf of Mexico, USA (e.g. Roberson & Lahann, 1981), and elsewhere (Pusch & Karnland, 1988). These studies suggest the illitisation rate in the natural environment is considerably slower than that predicted by kinetic models, but this is due to the fact that the process depends on the rate of supply of potassium, which may often be limited (as will also be the case at Olkiluoto).

However, these studies do not really represent the repository environment because the duration of heating is several orders of magnitude longer than would be the case in Olkiluoto (cf. Figure 1).

Conclusions and proposed study

NA studies of thermal alteration of bentonite are legion (and only a few are noted above), but critical review of the work carried out to date indicates that they are of little value in supporting the safety case because either the conditions are non-repository relevant or the information obtained is limited due to:

- inappropriate temperature range examined (e.g. Ishrini)
- inappropriate timescales for heating of the bentonite, they are usually way too long (e.g. Gulf of Mexico)
- ill-defined (Pellegrini et al., 1999) or assumed (e.g. Busachi) boundary conditions
- under-characterised physico-chemical parameters of the bentonite
    - use of modern analytical techniques can better define temperature zonation before assessing changes in the smectite (cf. comments in Posiva, 2013b)
    - define more than just the smectite content (e.g. swelling pressure, saturation state etc; cf. Alexander & Milodowski, 2015)

In addition, certain aspects have never been properly addressed, for example:

- as surface/near-surface sites are normally studied, bentonite under an Olkiluoto-relevant lithostatic load/hydralulic pressure has not been examined
- again, as surface/near-surface sites are normally studied, fully saturated bentonite has never been examined

Suitable sites have been identified for a new project on the thermal alteration of bentonite (TAB) and these will be discussed in the presentation as will overcoming the various uncertainties noted above. Now, with the new safety case for Olkiluoto about to be launched, isn't it time we got it right?

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