Subject category: 12
Target category: B

Hyperalkaline Natural Analogue Potential in the Philippines

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1. Introduction

Bentonite is one of the most safety-critical components of the engineered barrier system in the disposal concepts developed for many types of radioactive waste. The choice of bentonite results from its favourable properties (including plasticity, swelling capacity, colloid filtration, low hydraulic conductivity, high retardation of key radionuclides) and its stability in relevant geological environments (see Alexander & McKinley, 1999, for details). However, bentonite is unstable at high pH (e.g. Metcalfe & Walker, 2004). Due to the fact that cementitious materials react with groundwater to produce initial leachates with pH >13 (later falling to around pH 12.5), this led to some repository designs (e.g. Nagra, 2002), that specifically exclude the use of concrete in any sensitive areas containing bentonite. This has also driven recent interest in low alkali cements, because the pH of the leachate is somewhat lower than standard OPC (Ordinary Portland Cement), lying around pH 10-11. It is hoped that this lower pH will reduce bentonite degradation, so allowing the use of low alkali cements in close proximity with bentonite.

Assuring the long-termed stability of bentonite in contact with such hyperalkaline fluids under conditions representative of a deep geological repository for HLW or long-lived ILW requires complementary laboratory, modelling and *in situ* studies. In particular, to build a robust safety case, it is important to have supporting natural analogue data to confirm understanding – and validate models – of the long-termed performance of bentonite (Miller et al., 2000).

Natural analogue studies could:

- Provide quantitative information on alteration rates, the products of such alteration and their safety-relevance to the performance of the engineered barrier system
- Allow testing of current models and databases used to assess such alteration
- Input in technical aspects of the requirement to provide a range of supporting documentsfor safety cases

As a result of a review of the available literature and geological investigation (McKinley et al., 2008; Yamakawa et al., 2008), several sites in the Philippines were selected as particularly promising for this purpose; preliminary field investigations confirmed the presence of hyperalkaline springs with a pH of 10-12. Indeed, the studies indicated that on-going serpentinisation in the target area in the Zambales ophiolite (northwest Luzon) may have produced more pervasive high pH groundwater than any other location worldwide. Bentonite is mined in the vicinity and smectite layers are common, occurring in layers many hundreds of metres thick around local volcanic plugs. Bentonite/zeolite layers act regionally as aquitards, isolating flow of deeper high pH waters from surface runoff. Several sites are now under active investigation in the search for evidence of hyperalkaline groundwater/bentonite interaction and this programme has been developed into the International Philippines Hyperalkaline Analogue Project (IPHAP).

2. Natural analogue concept

Although systems representative of leachates from both OPC (i.e. groundwaters with pH 12.5 and above) and low alkali cements (groundwater pH of 10 to 11) have previously been examined as natural analogues of cementitious repositories (e.g. Bath et al., 1988; Khoury et al., 1992), neither produced data of relevance to the questions now being posed with regard to low alkali cements. The natural cements in Jordan are closely representative of OPC-based materials (Pitty, 2008; Alexander et al., 2008), so the pH is too high. An appropriate low alkali water chemistry was studied in Oman (McKinley et al., 1987, 1988), but this project did not include any investigation of mineral alteration.

An initial literature study was sponsored by METI[1] of Japan with search parameters that include both aspects of the target geology and also themes of relevance to radwaste management programmes. Factors considered include:

[1] This study was initiated within a project to develop an integrated natural analogue programme in Japan, which was funded by the Ministry of Economy, Trade and Industry, Japan (METI) and coordinated by RWMC.

- ophiolite terrains
- hyperalkaline groundwaters
- bentonites
- H_2 or CH_4 gas in groundwaters
- thermal groundwaters
- coastal sites
- logistics (e.g. potential support from local mining operations, ease of transport etc)
- potential for training junior staff.

This indicated that no useful information on this topic could be "mined" from any past studies and hence the option of a new project was examined. The basic idea was to use a "top-down" approach to identify sites where bentonite deposits have been exposed to relevant hyperalkaline water for very long periods. Especially given the interest in low alkali (and thus lower pH) cements, the focus was on sites that have natural waters with pH in the appropriate range (around 10-11). The cement leachate is simulated by natural hyperalkaline water, which, if the timescale of interaction can be determined, allows the models that are being developed to quantify the specific processes of relevance to bentonite alteration to be tested.

The challenge is to maximise the value of this test, by assuring that materials and boundary conditions are as similar as possible to those in a repository. Nevertheless, it must be emphasised that such sites are no more than an analogy of a repository, not a copy, and hence certain differences are inevitable. Currently, the technical focus is on:

- long-term bentonite stability in contact with water analogous to low alkali cement leachates
- if possible, same system as above interacting with seawater/brines for a coastal repository
- if possible, same system as above interacting with a range of leachate chemistries (cf. Table I) as the precise situation in a repository will depend both on the site conditions and the composition of the cementitious materials – neither of which have been fixed as yet
- low alkali cement leachate/host rock interaction – is there any?
- BPM (blind predictive modelling – see Pate et al., 1994) of the chemistry of safety relevant elements (eg Se), including *in situ* speciation
- colloid filtration
- microbiology of the system (cf. McKinley et al., 1988)
- staff training, including mentoring by experienced (in radioactive waste disposal) international staff

3. Overview of the results of the field programme

Ophiolites were identified as potential sources of relevant hyperalkaline water. The term ophiolite was originally used by Brongniart (1813) for an assemblage of green rocks (serpentine, diabase) in the French Alps. Ophiolites, as defined here, adapt the nomenclature of the Penrose Conference of 1972 (Geotimes, 1972) and, from top to bottom, consists of:

deep (abyssal) marine sediments
pillow lavas (basalt)
sheeted dyke complex
high level/isotropic gabbro
layered mafic cumulates (gabbro)
layered ultramafic cumulates
transition zone dunites and residual peridotites

If any of the above lithologies is missing, it should be called an ophiolite *complex*, but this term is frequently misused in the literature. There are a number of locations worldwide where such an analogue might be found, including Japan, Cyprus, Oman, Western USA, Bosnia-Herzegovina, Papua New Guinea and the Philippines. Based on a multi-attribute analysis, considering factors such as probability of finding suitable locations, relevance to Asian radioactive waste disposal programmes, opportunity for training, low risk of disrupting calls for volunteer sites in those Asian countries actively carrying out site characterisation programmes (e.g. Japan, Korea, China) and cost-effectiveness, the Philippines is now the preferred option for the Japanese programme and has recently been the focus of more detailed literature studies and a limited number of field investigations to confirm fundamental feasibility.

The target hyperalkaline pH waters (generally between pH 10 and 11, see Barnes & O'Neill, 1969 for examples) are a product of the serpentinisation of ultramafic rock. This reaction may occur by several possible pathways (e.g. Abrajano et al., 1988; Sader et al., 2007) with the exact reaction pathway depending on Mg content of the precursor olivine/pyroxene or serpentine product, CO_2 fugacity, water-rock ratio, Ca^{2+} content of groundwater, etc.

The serpentinite mineral assemblages are very strongly reducing and the hyperalkaline waters are often effervescent with H_2 and/or CH_4 gas; geochemical modelling (e.g. Neal & Stanger, 1983) and experimental evidence (e.g. Wright & Catlow, 1996) suggest abiogenic reduction during the serpentinisation process. Such reducing water is a potential energy source and springs can often be identified by characteristic microbial mats. Some of the reaction pathways are also strongly exothermic, frequently producing hydrothermal groundwaters, which are often used as therapeutic springs in the Philippines.

Several ophiolites on the islands of Panay (around E121,000 N012,500), Luzon (around E120,500 N014,500) and Palawan (around E118,000 N010,000) have been examined as part of a preliminary assessment of suitable sites. To date, the area around Brooke's Point in southern Palawan appears to be an excellent analogy for a coastal repository, with clear evidence of mixing of ophiolite-derived hyperalkaline groundwaters and marine-derived groundwaters under the coastal plain.

Logistically, the Mangatarem area (E120,180 N015,047) in the province of Pangasinan on the west central area of the island of Luzon in northern Philippines appears the most promising. This area is part of the Eocene Zambales ophiolite (Figure 1), which consists of three volcanic-hypabyssal units (Yumul and Dimalanta, 1997): the Coto Block volcanic-hypabyssal rocks, the Coto dykes and the Acoje Block volcanic-hypabyssal rocks – with the Cote Block being of most relevance to this project. A significant body of work exists on the Zambales ophiolite (e.g. Rossman et al., 1989), but most is focussed on the northwestern Acoje Block, no doubt prompted in part by the rich mineral reserves in the area. For example, according to Sturchio et al. (1989), serpentinisation of the rocks of the Acoje Block occurred at temperatures between 30 and 350°C, but no data are available for the Cote Block for comparison. Samples have been collected from the pillow lavas, both near the Manleluag Springs and further west and south, to assess this for the Mangatarem area (see Figure 2) and to evaluate the serpentinisation pathway. In addition, samples have been collected to assess the uplift history of the site, so as to constrain the likely period of leachate/bentonite interaction.

This area also hosts the largest known bentonite deposits in the country (the Saile Industries quarry: see Figure 2), with reserves estimated to be approximately four million tonnes (Ugalde, 1999). Here, bedded bentonites and zeolites belong to the Eocene Aksitero Formation (and most probably extend to the finer members of the overlying Moriones Formation), the sedimentary carapace of the Zambales ophiolite unconformably which conformably overlie the cherts and upper pillow lavas of the ophiolite. Examination of the Aksitero Formation in a range of localities (including the type locality at Bigbiga and in the immediate vicinity of the Mangatarem quarry) make it quite clear that the relationship is conformable. For example, basal limestones were observed filling spaces in and around the pillow lavas and breccia and penetrating up to several metres into fractures and cooling joints. In places, bentonitic clays can be observed lying directly on the basalts, for example at the Formation type section in the Aksitero River valley, south of Bigbiga (see Figure 2 in Schweller et al., 1984). Very localised repetition of pillow lavas above the bentonite were observed by the authors, clearly showing the contemporaneous and conformable nature of the relationship between the bentonite and the underlying ophiolite.

Although exposure is generally poor, the confirmed extent of the formation is some 35km, north to south, and several hundred metres east to west. Moving from the base of the Aksitero Formation and up into the unconformably overlying Moriones Formation, it is possible to follow the onset of deep (reportedly >1000 m, but certainly less than the calcite compensation depth), marginal basin-sedimentation on the recently formed pillow lavas through a slow decrease in water depth (and move towards shallow water deposits including clasts of the ophiolite, wood, corals etc) as the ophiolite is uplifted and exposed (on accretion at the Manila Trench), leading to the current situation of erosion of the ophiolite contributing to infilling the Central Valley of Luzon to the east. It would be useful to have an estimate of the rate of uplift on the ophiolite to constrain the onset of leachate/bentonite interaction. Estimates exist in the literature (e.g. Encarnación, 2004) and these could be checked by examination of some of the gabbro samples collected around the Manleluag Spring.

The well-bedded tuffaceous members of the Aksitero Formation have been authigenically transformed after deposition into bedded zeolite and a bentonite[2] dominated by montmorillonite (Ca-smectite) and quartz which is similar in bulk chemistry to the MX-80 and Kunigel V1 bentonites which are under consideration as an EBS material in many national programmes (see Table 2). Individual beds range in thickness from a few centimetres to several metres and range in colour from light cream, beige, off-white, light to medium brown and greenish. Texture closely resembles a tuff, but sometimes exhibits finer grains, and sparse Mn-nodules can be found disseminated through the bentonite at some sites.

In the immediate vicinity of the bentonite deposit, hyperalkaline groundwaters from the sheeted dykes and pillow lavas of the ophiolite come to the surface at the Manleluag Hot Springs, some 800-900m to the southwest of the quarry,. The groundwater chemistry is clearly that of a serpentinisation-derived (presumably from the host ophiolite) groundwater which has close similarities to low alkali cement leachates (see Table 1). In addition to the several known springs around the Manleluag site, a pervasive presence of hyperalkaline waters can be detected in stream surveys and there is clear evidence that a N50E trending fault system is controlling the hyperalkaline groundwater distribution in the area.

The bentonite appears (logically) to be acting as an aquiclude, in that the hyperalkaline groundwaters can only be found beneath the bentonite when it is present, with lower pH groundwaters found on top of the bentonite. This is relevant as the bentonite here is predominantly in the Ca-form, which could represent an analogue of the first, rapid ion-exchange alteration of an initial repository Na-bentonite exposed to low alkali cement leachate. Observations indicate that such a process does not seem to compromise its hydraulic barrier role.

Although hyperalkaline leachate/bentonite interaction has not yet been definitely identified (this awaits reporting of the analysis of appropriate bentonite samples), the potential for reaction in this area is very high as the N50E trending fault system runs under the bentonites of the Aksitero Formation and appears to have induced some fracturing in the bentonite itself (Figure 3). Samples of fracture-filling material have been

[2] In addition, in the northern part of the quarry, a 1m thick bed of talc has been tentatively identified, perhaps representing signature of high pH alteration in a high Mg system groundwater.

collected from the quarry for analysis. The presence of Mn-oxide coatings on the fractures, suggests that the hyperalkaline groundwaters have, in the past, been channelled along these fractures although there is no evidence of any flow at present. More extensive Mn-staining of exposures at deeper levels of the mine indicates also more general, slow transport (diffusion dominated?) from an underlying Mn source, presumably dissolution of Mn nodules, which may be facilitated by the high pH.

In addition, there is good evidence that other safety assessment relevant work on gas and colloid transport, microbiology and radionuclide solubility/speciation could be conducted at this site.

4. Conceptual model of the Mangatarem site

Following exploratory field trips to several ophiolites in the Philippines, work has most recently been focussed on the Mangatarem area of Luzon. Observations from this area have been supported by information from other sites to produce an integrated conceptual model for Mangatarem in particular, but also for the other ophiolite-hosted hyperalkaline systems observed in and around the Zambales ophiolite (e.g. at Botolan, see Table 1). Here, the active serpentinisation processes in the ophiolite (D in Figure 4) are driving the production of heated, gas-rich hyperalkaline groundwaters. Transport through the ophiolite is fault controlled and either comes to the surface (e.g. A, Figure 4) where the bentonite cover is absent or is effectively trapped under the bentonite aquiclude (C, Figure 4) where this is present. The hyperalkaline groundwaters below the bentonite have been observed in deeper drillholes (e.g. in the Bigbiga area to the south of Mangatarem) and it is assumed that the base of the bentonite will be a rich zone of hyperalkaline interaction (E, Figure 4). In addition, the groundwaters appear to have come up through the bentonite where it has been fractured by regional tectonic forces (B, Figure 4).

Mangatarem is the focus of the next field campaign in autumn 2008 where sampling is intended to provide clear evidence of hyperalkaline groundwater/bentonite leaching. In addition, groundwater samples will be collected to assess buffering reactions by the bentonite as will drill samples from the ophiolite in an attempt to assess the uplift history of the ophiolite and the likely serpentinisation pathway(s)[3].

5. Conclusions

Preliminary field surveys in the Philippines have identified three major ophiolite / bentonite localities which offer several environments of direct relevance to a range of radioactive waste disposal programmes. Further work at these sites offers a high probability of success in meeting the main project goals. Recently, the focus has been on the bentonites of the Zambales ophiolite which have provided:

- at least two good sites in close vicinity (Mangatarem and Bigbiga); the former has strong local support from the bentonite mining company and offers good infrastructure
- indirect evidence collected so far indicates ongoing bentonite/hyperalkaline interaction and this will be verified later this year following analysis of appropriate bentonite samples
- a further good backup site (Poon Bato, on the southwest of the Zambales ophiolite) has a particularly wide distribution of hyperalkaline waters in case this turns out to be problematic at the priority locations

In addition, there are very good chances of obtaining several of the project secondary goals if sufficient funding is available to extend investigations. Overall, the project would be well suited for international collaboration – particularly for partners in East Asia – as it offers an opportunity for staff to work in a complex project involving aspects of site characterisation, repository design and performance assessment, which provides invaluable experience for future repository implementation.

6. Acknowledgements

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[3] Note that seven potential serpentinisation reaction pathways are known, so it is likely to be a complex picture.

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Table 1: Hydrochemistry of hyperalkaline groundwaters: examples from around the world for comparison with the novel Philippines groundwater data and low pH cement leachates. All data in ppm (nd: no data bdl: below detection limit).

Location	pH	Na	K	Ca	Mg	Cl	SO_4	SiO_2
Philippines[1]								
Manleluag 1	11.1	28.0	0.5	18.6	0.2	17.4	5.1	nd
Manleluag 2	10.4	20.6	0.4	18.1	nd	15.8	nd	21.1
Poon Bato	10.9	18.4	0.9	33.1	0.05	20.9	nd	2.5
Narra 1	10.8	158	0.9	3.1	0.0	95.0	nd	nd
Narra 3	10.3	157	0.9	2.4	0.1	80.0	bdl	44.2
Worldwide[2]								
Cyprus 3a	11.5	385	15.1	1.0	0.3	420.0	251.0	24.0
Greece	11.3	24.0	1.0	34.0	0.3	15.0	3.0	2.0
Bosnia	11.7	35.0	1.5	29.0	7.0	20.0	2.0	0.9
Oman	11.5	132	4.8	34.0	1.3	127.5	22.5	3.0
New Caledonia	10.8	15.0	3.0	14.0	2.3	22.0	0.8	0.4
Western USA	11.5	19.0	1.0	40.0	0.3	63.0	0.4	0.4
Cement								

leachate[3]								
ALL-MR f63	11.0	42	7.3	20	<0.5	52	12	49.2
OL-SR f63	10.0	4400	150	4300	0.56	13000	247	32.1

1: this project 2: Neal & Shand, 2002 3: Vuorinen et al., 2005

Table 2: Geochemical (XRF) analysis of the Mangatarem bentonite and MX-80 bentonite (in % by weight) and cation exchange capacity (CEC)

Parameter (%)	**Untreated Mangatarem**	**Average treated Kunigel V1 (Nakashima, 2004)**	**Average treated MX-80 (Wyoming) (Karnland et al., 2006)**
SiO_2	50.3 - 69.8	71	58.5
TiO_2	0.40 – 0.64	0.20	nd
Fe_2O_3	2.8 – 6.8	2.1	3.8
Al_2O_3	12.1 - 13.2	14	19.1
CaO	0.1 – 5.8	2.3	1.4
MgO	1.9 - 3.1	2.3	2.4
K_2O	0.4 - 1.2	0.33	0.5
Na_2O	1.2 - 1.3	2.6	2.1
CEC	67.5 meq/100 g[4]	56 meq/100 g	72.0 meq/100 g

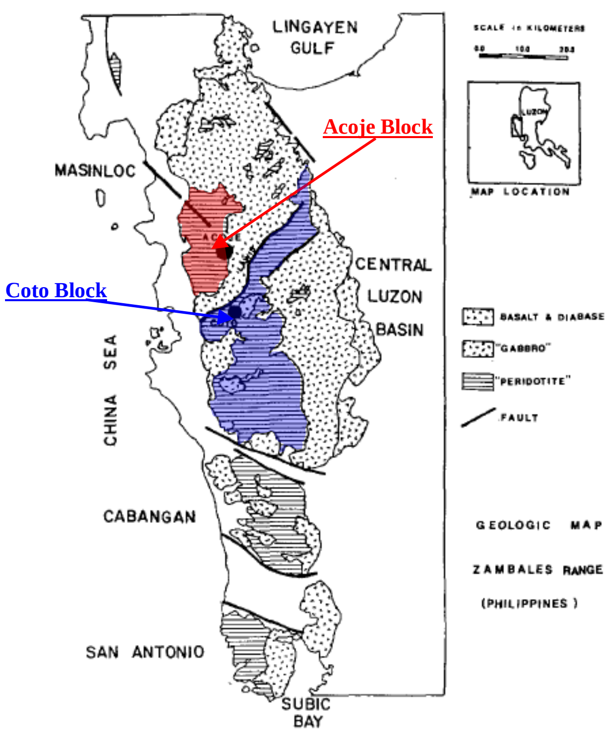

Figure 1: Regional geological map of the Zambales ophiolite, showing the Acoje Block in the northwest. The Cote Block is to the south of this, separated by a major fault line. From Abrajano and Pasteris (1989).

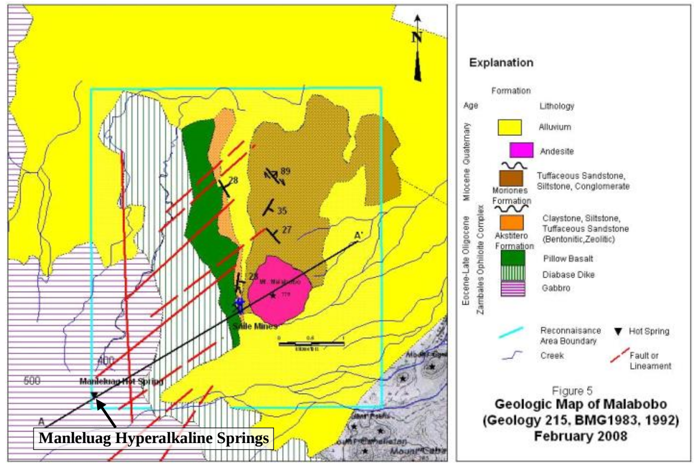

Figure 2: Geological map of the main study area at Mangatarem, northwest Luzon, Philippines. The Manleluag Hyperalkaline Springs are just off the map to the southwest.

Figure 3: Reverse faulting in the bentonite at the Saile quarry. Compass case in picture is approximately 8 cm across.

[4] From Fernandez, 1999.

Subject category: 12
Target category: B

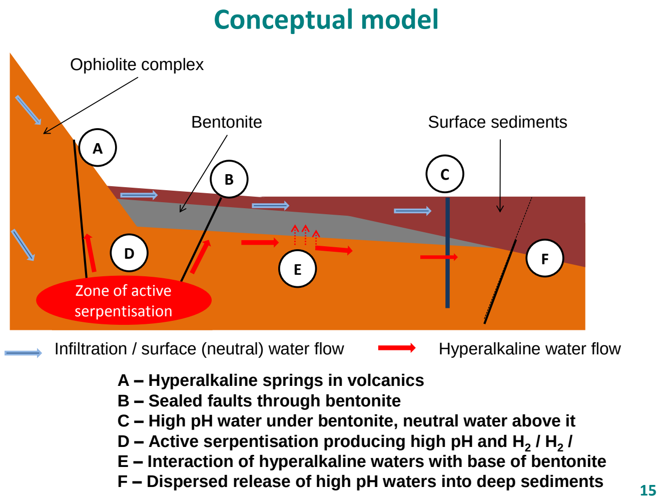

Figure 4: Conceptual model of the hyperalkaline groundwater/bentonite interaction in the Zambales ophiolite area