Bedrock Geosciences Technical Report 2023-02

Proceedings of the NAWG-16 Workshop
15-18th October, 2019, Zao, Yamagata, Japan

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1. Introduction

1.1 Overview of the workshop

The 16th Workshop of the Natural Analogue Working group (NAWG) was held in the Hotel Jurin in the mountain hot spring village of Zao Onsen in Yamagata Prefecture in Japan (see Figure 1-1). The workshop ran from Tuesday, 15th to Friday, 18th October, 2019 and consisted of 2.5 days of presentations and discussion followed by 1.5 days of field trips (see Table 1-1).

Figure 1-1: View of Zao Onsen, Yamagata Prefecture, Japan

Table 1-1: NAWG-16 Workshop Agenda

Monday 14th October		
	Travel to the venue	
18:00 – 19:00	Registration	
19:00 – 21:00	Icebreaker and dinner in the Yamagata Sake Museum	
Tuesday 15th October		
09:00 – 09:10	Opening of the workshop and organisational aspects	Russell Alexander Chairman NAWG
09:10 – 09:20	01_Welcome from NUMO	Motoyuki Yamada NUMO, Japan
09:20 – 10:00	02_Current status of the Japanese national programme	Kunio Ota NUMO, Japan
Session Ia	**Near-field studies**	**Chair: Timo**
10:00 – 10:30	03_Bentonite analogue research for spent nuclear fuel disposal (SNFD) in Taiwan	Polly Tsai

		INER, Taiwan
10:30 – 11:00	*Coffee/tea*	
Session Ib	**Mini-session on archaeological analogue studies**	
11:00 – 11:30	04_Overview of the Czech programme of archaeological analogues for long-term canister lifetime prediction	Jan Stoulil University of Chemistry and Technology Prague, Czech Republic
11:30 – 12:00	05_Archaeological analogue studies at JAEA	Seiichiro Mitsui JAEA, Japan
12:00 – 12:15	06_Comparison with the results of the Cyprus NA Project	Russell Alexander Bedrock Geosciences, Switzerland
12 :15 – 13:45	*Lunch*	
Session III	**Far-field studies**	**Chair: Kunio**
13:45 – 14 :15	07_Palaeohydrogeological evolution of the Horonobe site, Sea of Japan	Isao Machida AIST, Japan
Session Ic	**Mini-session on PNAP – low alkali fluid reaction with bentonite**	
14 :15 – 14 :45	08_The Philippines International Analogue Project: an overview of the project	Naoki Fujii RWMC, Japan
14 :45 – 15 :15	09_Formation of metastable phases and transformation to stable phases under alkaline condition in Palawan, Philippines	Misato Shimbashi CRIEPI, Japan
15:15 - 15:45	*Coffee/tea*	
15:45 – 16:15	10_Bentonite-alkaline fluid interaction at bentonite deposit in the Philippines: pore clogging by secondary precipitates formed by the interaction	Tom Sato Hokkaido University, Japan
16:15 – 16:45	*11_Application of non-destructive methods in bentonite analogue research at GTK*	*Heini Reijonen*

		GTK, Finland
16:45 – 17:00	Discussion	
Session II	**Posters session from 17:00**	
Dinner	Hotel Jurin	
Wednesday 16th October		
Session III	**Far-field studies I**	**Chair: Russell**
09:00 – 09:30	12_Pre- and postglacial palaeoseismicity in Finland – evidence of multiple bedrock fault reactivation events	Timo Ruskeeniemi GTK, Finland
09:30 – 10:00	13_Matrix diffusion in the geosphere: the UK perspective	Simon Norris RWM, UK
10:00 – 10:30	*Coffee/tea*	
10:30 – 11:00	14_The use of the Greenland Analogue Project (GAP) in the Finnish Safety Case	Nuria Marcos AINS, Finland
11:00 – 11:30	15_Bedrock fracturing – characteristics and prediction at different scales. A self-analogue for the Finnish repository site	Jon Engström GTK, Finland
11:30 – 11:45	16_A natural analogue study in KAERI: geochemical characteristics of deep geology in KURT	Ji-Hun Ryu KAERI, ROK
11:45 – 13:15	*Lunch*	
Session IV	**Far-field studies II – self-analogues**	**Chair: Simon**
13:15 – 13:30	17_Use of self-analogues in the Japanese SC	Saori Yamada NUMO, Japan
13:30 – 14:00	18_Long-term stability of fracture systems and their behaviour as flow paths in crystalline rock: an overview	Dora Yoshida Nagoya University, Japan
14:00 – 14:15	19_Introduction to the KINA (Kiruna International Natural Analogue) project	Russell Alexander

		Bedrock Geosciences, Switzerland
Session V	**The use of NA in stakeholder communications**	**Chair: Venda**
14:15 – 14:45	*20_Perception and communication of uncertainties by/with stakeholders: overview of the recent NEA meeting on Managing Uncertainty in Repository Siting and Implementation – Creating a Dialogue between Science and Society*	*Ulrich Noseck* *GRS, Germany*
14:45 – 15:30	*Coffee/tea*	
15:30 – 16:00	21_Communication with NA in Japan	Ken Kaku NUMO, Japan
NAWG-16 Workshop Dinner 19:00	Hotel Jurin	
	Thursday 17th October	
Session V (cont.)	**The use of NA in stakeholder communications**	**Chair: Venda**
09:00 – 09:30	*22_The use of natural analogues in building stakeholder confidence in Canada's plan for the long-term management of used nuclear fuel*	*Erik Kremer* *NWMO, Canada*
Session VI	**Microbiology in the bentonite buffer**	**Chair: Dora**
09:30 – 10:00	*23_Microbial populations in natural bentonites: results from IBL*	*Erik Kremer* *NWMO, Canada*
10:00 – 10:30	24_Biotransformation of actinides and fission products	Toshi Ohnuki Tokyo Institute of Technology, Japan
10:30 – 11:00	25_Microbial population assessment in bentonites from URL experiments in Japan	Yuki Amano JAEA, Japan
11:00 – 11:15	*Coffee/tea*	
Session VII	**Introduction to the International Bentonite Longevity (IBL) project**	**Chair: Ken**
11:15 – 11:45	26_International Bentonite Longevity (IBL) project: an introduction (Description of the of the Tsukinuno bentonite mine)	Russell Alexander Bedrock Geosciences, Switzerland

11:45 – 12:00	27_Description of the of the Dobuyama bentonite quarry	Masakazu Ito Kunimine Industries, Japan
Session V (cont.)	**The use of NA in stakeholder communications**	
12:00 – 12:15	*28_Stakeholder communication with natural analogues: an international perspective*	*Dorine Montout* *Univ. Paris 1 Pantheon-Sorbonne, France*
12:15 – 13:00	*Lunch*	
Session VIII	**The influence of geology on the local Yamagata wine**	**Guide: Saori**
	29_Introduction to Asahimachi Winery	Saori Yamada NUMO, Japan
13:30	Travel to the Asahimachi Winery	
14:30	Meet to examine the influence of geology on the local wine (with a tasting and vineyard visit) at the Asahimachi Winery	
16:00	Travel to the Route Inn hotel in Yamagata city	
17:00	Free evening	
Friday 18th October		
Session IX **(09:00 – 17:00)**	**Field trip to Kunimine Industry Company's bentonite quarry at Dobuyama and outcrops around their bentonite mine at Tsukinuno**	
ca. 17:00	End of workshop, return to central Yamagata for onward travel	

Note that Taifun Hagibis, the most powerful to hit Japan in 2019, made landfall just before the start of NAWG-16 and this caused disruption to flights into Japan, meaning several participants (Table 1-2, *in italics*) missed the workshop. Luckily, several of them could still make their presentations via online links and these are marked in Table 1-1 *in italics*. Disruptions in Japan also meant last minute changes to plans for some participants and this is also reflected in the somewhat confused order of presentations in Table 1-2.

The workshop agenda consisted of several sessions with a mix of natural analogue (NA) topics of relevance to radioactive waste disposal:

- Near-field studies, including:
    - Analogues of various waste forms
    - Canister studies (including a special sub-session on archaeological analogues in Japan)
    - Buffer and backfill/plug and seal analogues (including a special sub-session on RWMC's recently completed Philippines bentonite study)
    - Long-term impact of microbial processes
- Far-field studies, including:
    - Radionuclide retardation in the geosphere
- Recent examples of NA support of the safety case (SC)
- Stakeholder communication with NAs

The two, bespoke, field trips covered:

- an introduction to the International Bentonite Longevity project (IBL – please see www.iblproject.com) with visits to Dobuyama bentonite quarry of Kunimine Industries Company (KIC) and their Tsukinuno bentonite mine, focus of IBL
- examining the influence of geology on the local wine (with wine tasting and vineyard visit) at Asahimachi winery, Yamagata Prefecture

The participants (Table 1-2 and Figure 1-2) came from a range of organisations from some ten countries worldwide.

Table 1-2: List of workshop participants (*in italics* means the individual was not physically present at the meeting, see explanation above)

Participants name	Organisation and country
1. Ulrich Noseck	*GRS, D*
2. Andree Lommerzheim	*BGE, D*
3. Heini Reijonen	*GTK, FI*
4. Timo Ruskeeniemi	GTK, FI
5. Kunio Ota	NUMO, J
6. Saori Yamada	NUMO, J
7. Venda Havlova	UJV, CZ
8. Vlastislav Kašpar	UJV, CZ
9. Jan Stoulil	University of Chemistry and Technology Prague, CZ
10. Dorine Montout	*Univ. Paris 1 Pantheon-Sorbonne, F*
11. Dora Yoshida	Univ. Nagoya, J
12. Yuki Amano	JAEA, J
13. Tom Sato	Univ. Hokkaido, J
14. N.Fujii	RWMC, J
15. M.Shimbashi	CRIEPI, J
16. Seiichiro Mitsui	JAEA, J
17. Simon Norris	RWM, UK
18. Erik Kremer	*NWMO, C*
19. Masakazu Ito	KIC, J
20. Nuria Marcos	AINS, FI
21. Isao Machida	AIST, J
22. Jon Engström	GTK, FI

23. Polly Tsai	ITER, ROC
24. J-H.Ryu	KAERI, ROK
25. Bill Miller	Wood plc, J (now Jacobs, J)
26. Mika Yamada	Wood plc, J (now Jacobs, J)
27. Toshi Ohnuki	Tokyo Institute of Technology, J
28. Yuto Nishiki	Univ. Hokkaido, J
29. Ryosuke Kikuchi	Univ. Hokkaido, J
30. Russell Alexander	Bedrock Geosciences, CH

Figure 1-2: *NAWG-16 participants (image courtesy K.Ota, JAEA, Japan).*

1.2 Why natural analogues - assessing repository performance at long times?

A major challenge in the development of a SC for a deep geological repository is dealing with the long period of time over which the wastes remain hazardous. Over such a period, a wide range of events and processes acts on a repository and it's geological and surface environment. These events and processes, taking place over different time windows and at local to regional scales, result in increasing uncertainty in the future evolution of the repository and its environment. This means that arguments must be developed to show that this uncertainty can be addressed in a manner that is not only acceptable to regulators, who may in any case set out the types of arguments they want to see, but also convincing to less technical audiences who need to trust in the safety of the repository. Thus complementary lines of argument are required to compensate for increasing uncertainties affecting calculated releases at distant times.

However, complementary arguments[1] can also be made to address other aspects of safety, especially continuing isolation of the wastes, even at times beyond when quantitative safety assessments (SA) can be supported. NEA (2009) suggests that "complementary arguments might be based, for example, on the absence of resources that could attract inadvertent human intrusion and on the geological stability of the site, with low rates of uplift and erosion". Another challenge with the long period addressed by the SC is that, although some experiments can be carried out in the laboratory, in underground research facilities (thus in the actual or similar host rock and geological environment) or in the field, these cover short timescales compared with long-term repository evolution. To try and address this specific problem, complementary arguments are made using analogous geological and/or anthropogenic examples of the materials and processes of interest (see the discussion on NA, below) to show that understanding is good enough to extrapolate short-term experimental results to long-term performance.

A further challenge arising from the long time periods of interest in the SC relates to how safety is quantified over these very long times. The most common indicators of safety are individual dose and risk (NEA 2002) and, of these, dose is much easier to communicate to a wider audience as it can be compared, for example, with the natural background radiation or medical radiation exposures (comparisons which are themselves complementary arguments).

Within the SC, quantitative SA using models and data tends to focus on potential radionuclide releases from a repository to the biosphere or surface environment. The uncertainties affecting the models can generally be quantified or bounded and dealt with in the SA by using cautiously chosen parameter values, conservative model assumptions or evaluating multiple cases covering the ranges of uncertainty. However, where the consequences of calculated releases are to be expressed in terms of dose, the biosphere must also be modelled. The models of the way in which humans are exposed (e.g. ingestion via consumption of food or drinking of water) are closely related to human habits that can be predicted with confidence only in the very short term, basically in the order of decades.

To complement the quantitative estimates of doses, especially in the period beyond a few tens of thousands of years, additional complementary safety indicators have been proposed (e.g. IAEA 2003) using fluxes and concentrations of naturally-occurring radionuclides in the undisturbed biosphere or geosphere for comparison with the calculated radionuclide releases from the repository. IAEA (2003) also found that alternative indicators such as "crossover times" could be useful in illustrating safety. A crossover time is the point in time in the future at which either the activity or radiotoxicity of the radionuclides remaining in the engineered barriers or released to the geosphere decrease due to radioactive decay below the corresponding values for relevant natural materials such as the original uranium ore or the excavated host rock. Both these areas were explored in Alexander et al (2015a).

1.3 Introduction to natural analogues – what are they and how do they help us assess safety?

The main arguments employed here relate to NAs of the repository systems or processes. As noted in Miller et al. (1994, 2000), argumentation by use of analogy is well established in many fields including philosophy, biology, linguistics and law (Petit, 1992), and most scientists are familiar with

[1] Also termed 'complementary considerations', see Posiva (2013a, 2023)

this approach and will have used it at some point in their career. For example, in the oil industry, accessible (surface) analogues of the geological conditions expected in physically inaccessible deep oil and gas reservoirs are often studied.

For the specific case of radioactive waste disposal, the main inaccessible features are:

- the very long time it will take for long lived waste to decay to safe levels – how can anyone know how the materials which are used to contain the wastes will behave over thousands to millions of years?
- the large spatial scales which cannot be directly addressed in a laboratory – how can the migration of radionuclides through several hundred metres of host rock from the repository to the earth's surface be studied and modelled?
- the heterogeneity and structural complexity of the geological environment which will host the repository – how can this ever be approached in a laboratory or modelled on a computer?

Hence the study of natural (predominantly geological) systems has been termed natural analogue research within the radioactive waste disposal community and the term "NA" has developed a particular meaning associated with providing supporting arguments for a repository SC (see, for example, Chapman et al. 1984; Côme & Chapman 1986; Miller et al. 1994, 2000; Posiva 2013a; Alexander et al. 2014, for discussion). As noted above, the key factors here are the heterogeneity and complexity of natural systems and, in particular, the very large dimensions and long timescales over which safety must be assured.

Due to the long timescales of concern, the basis of most SCs is a quantitative evaluation that is based on complex mathematical models and their general lack of transparency only adds to the mistrust of many stakeholders. How then can people be convinced that it is possible to assess the performance (and thus ensure the safety) of a repository over the long timescales of interest? One way is to address the robustness of the SA models, by clearly indicating the form and extent of model testing carried out within the repository SA. Not only can this show that the individual component parts of the complex structure which constitutes most SA models have been checked, but also that the 'mathematical black boxes' (cf. Alexander et al. 2003) constitute an acceptable representation of the repository system.

As noted by Alexander et al. (1998), part of the problem undoubtedly lies in the unusual nature of radioactive waste disposal: in most major engineering projects, such as bridge construction or aerospace engineering, the designs are tested against a range of laboratory experiments backed up by expert judgement based on experience with the same or similar systems. Here repository design deviates from standard engineering practice in that only a few repositories currently exist and testing their compliance to design limits will be impossible due to the timescales involved. In addition, peoples' anxiety about most things radioactive means that they require some greater form of 'proof' that a repository is safe than they are willing to accept for other engineered systems (see discussion in West et al., 2002, for example). This being the case, significant additional effort has been expended within the radioactive waste disposal community to make it clear that the SA models can adequately predict the long-term behaviour of a repository.

1.4 What is a natural analogue?

Traditionally, SA modellers have placed much weight on laboratory data for the construction and testing of their SA models and, with only a few exceptions (e.g. Posiva, 2013b), have not integrated in their SA reports data from either NAs or in situ experiments in URLs (Underground Rock Laboratories) – see discussion in Alexander et al. (1998). The over-dependence on laboratory data is understandable in that the information is produced under well understood, fully controlled conditions and thus the modellers feel they can place a high degree of confidence in the results obtained. Unfortunately, the full complexity of a repository cannot be re-created in a laboratory and it is necessary to address processes which are influenced by natural heterogeneities, which include large degrees of uncertainty and which operate over very long timescales. In this case, it is necessary to supplement laboratory data with information from in situ URL experiments and NAs. The potential evolution of geological repositories can be simulated by the use of mathematical models, but the extent to which such models can be validated by conventional approaches is inherently limited. Here NAs have an unique role to play.

In its basic form, a NA study can be any form of investigation of any relevant natural system, as long as it provides quantitative (or even qualitative) information which can be used to support (and build confidence in) geological disposal. This may mean that a study provides data which are directly applicable to the SC or, alternatively, it may provide illustrations of concepts or processes which can demonstrate safety (see, for example, Reijonen et al., 2015, for discussion). Each repository design will require unique information to assist in building and presenting the SC but, historically, NA studies have tended to focus on only a narrow range of natural systems. They can thus be categorised into a few broad groups which are representative of some major components of a repository system or feature of its evolution, namely:

- natural geological and geochemical systems
- archaeological systems
- sites of anthropogenic contamination

It should be noted, however, that this focus is currently changing: Reijonen et al. (2015), for example, present an example of a more broad-based approach to the use of NA in supporting the SC whereas Baik et al. (2015) and Wolf & Noseck (2015) look to define specific forms of NA support for repositories in crystalline and evaporite host rocks, respectively.

It is beyond the scope of this report to provide an overview of the vast range of NAs that have been studied; for this the reader is invited to examine publications focussed on just that, such as Miller et al. (2000, 2006), CSN (2004), Degnan et al. (2005) and Brasser et al. (2008).

1.5 NAWG – the Natural Analogue Working Group

In an attempt to define NA studies more clearly, and to orient them towards individual processes for which good analogues can be found, Chapman et al. (1984) listed a set of guidelines for selecting NAs for investigation. The need for well-characterised, process-oriented NA studies is reaffirmed in this report and thus these guidelines are repeated here:

1) The process involved should be clear-cut. Other processes which may have been involved in the geochemical system should be identifiable and amenable to quantitative assessment as well, so that their effects can be subtracted.
2) The chemical analogy should be good. It is not always possible to study the behaviour of a mineral system, chemical element or isotope identical to that whose behaviour requires assessing. The limitations of this should be fully understood.
3) The magnitude of the various physico-chemical parameters involved (pressure, temperature, pH, Eh, concentration etc.) should be determinable, preferably by independent means and should not differ greatly from those envisaged in a repository.
4) The boundaries of the system should be identifiable (whether it is open or closed, and consequently how much material has been involved in the process being studied).

5) The timescale of the process must be measurable, since this factor is of the greatest significance for a natural analogue.

The quantitative philosophy outlined by Chapman et al. (1984) proved to be the impetus for a greater interest in natural analogues which resulted in an international symposium (Smellie, 1984) and the formation of the Natural Analogue Working Group (NAWG) which was originally sponsored by the Commission of the European Communities (CEC, now EC). Since its inception, these themes have been addressed at 16 international NAWG workshops (including NAWG-16, the focus of this report):

- 15th NAWG Workshop was held from Tuesday, 23rd to Thursday, 25th May, 2017 at the ÚJV laboratory, Řež, Prague in the Czech Republic. The main presentations at NAWG 15 are available at https://www.natural-analogues.com/nawg-workshops/nawg-15-2017-czech-republic
- 14th Workshop took place on 9-11th June, 2015 in Rauma, Finland. Here, the application of NAs to repository SA was discussed (see Alexander et al, 2015b for details)
- 13th Workshop took place on 13-16th May, 2013 in Nagoya, Japan. A wide area of NA study was covered and some 9 of the presentations were published in the Swiss Journal of Geosciences (volume 108, 2015)
- 12th Workshop took place on 11-13th May, 2011 in Larnaca, Cyprus where predominantly NAs of engineered barriers were discussed. The main presentations at NAWG 12 are available at https://www.natural-analogues.com/nawg-workshops/nawg-12-2011-cyprus
- 11th Workshop: Liverpool, UK, 2009 where a short meeting was held at the 12th ICEM (International Conference on Environmental Remediation). No proceedings were produced
- 10th Workshop: Munich, Germany, 2007 where the workshop examined how current and future studies could be better focussed on providing appropriate data for the various end-users of natural analogue data. The main presentations at NAWG 10 are available at https://www.natural-analogues.com/nawg-workshops/nawg-10-2007-germany
- 9th Workshop: Aarau, Switzerland, 2002, where a short meeting was held with the theme being the current international status of natural analogues. No proceedings were produced
- 8th Workshop: Strasbourg, France, 1999, was devoted to a presentation of three, major international natural analogue projects, Oklo (II), Palmottu and Pena Blanca (EUR 19118). See https://www.natural-analogues.com/nawg-workshops/other-nawg-workshops for a link to the proceedings
- 7th Workshop: Stein am Rhein, Switzerland, 1996, where one of the main themes of the workshop was the application of natural analogues to toxic wastes (EUR 17851 EN)
- 6th Workshop: Santa Fe, USA, 1994, where the intention was to review the "state-of-the-art" of several key issues in near-field and far-field processes and their importance to PA with the intention to provide a consensus view of the remaining areas requiring further research in natural analogues (EUR 16761). See https://www.natural-analogues.com/nawg-workshops/other-nawg-workshops for a link to the proceedings
- 5th Workshop: Toledo, Spain, 1992, was held in association with the final workshop of the Alligator Rivers Analogues Project (EUR 15176). See https://www.natural-analogues.com/nawg-workshops/other-nawg-workshops for a link to the proceedings
- 4th Workshop: Pitlochry, Scotland, 1990, was devoted to review 5 years of NA studies and the final conclusions drawn from the Poços de Caldas study (EUR 13014). See https://www.natural-analogues.com/nawg-workshops/other-nawg-workshops for a link to the proceedings
- 3rd Workshop: Snowbird, USA, 1988, where the application of natural analogues to repository performance assessment was discussed (EUR 11725)

- 2nd workshop: Interlaken, Switzerland, 1986, where anthropogenic analogues and the role of colloids, complexes and microbes have been reviewed (EUR 10671)
- 1st Workshop: Brussels, Belgium, 1985, where the theme was the interaction between the modellers and experimenters (EUR 10315). See https://www.natural-analogues.com/nawg-workshops/other-nawg-workshops for a link to the proceedings

NAWG reports are a particularly valuable record of the evolution and application of NA studies. Most of the large and significant analogues have been represented at NAWG meetings. Furthermore, the NAWG reports attempt to develop the perception of NAs by giving agreed introductory statements on their development.

Advances made since the formation of NAWG include:

- Studying NAs has greatly increased the understanding of repository-relevant processes and improved the capability to describe and effectively model them
- The larger, multi-objective NA studies can be a very cost effective way of training site characterisation and SA groups on real, complex systems (if properly integrated)
- The application of NAs in broadening public perception of the natural context of waste disposal has become increasingly important
- An increased awareness of the potential for studying NAs of chemo-toxic waste (e.g. Alexander & McKinley, 1999; Wilson et al., 2009) and understanding the long-term impacts of CO_2 sequestration (see, for example, Benson et al., 2005 and https://www.natural-analogues.com/nawg-library/ccs)

NAWG continues to grow (please see https://www.natural-analogues.com/background/nawg-members for details) and it is foreseen that the 17th workshop will be held in Biel/Bienne (Switzerland) at the end of November, 2021 and the 18th in Copenhagen at the end of August, 2022[2].

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[2] Unfortunately, the COVID pandemic meant that NAWG-17 will finally take place in May, 2023.

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2. Extended abstracts

2.1 Comment

Due to the unforeseen impact of the COVID-19 pandemic on project schedules, several authors have decided to withdraw their abstracts. It is hoped that some will be presented in an updated fashion at NAWG-17 (please see section 1.5, chapter 10 and www.natural-analogues.com for details).

2.2 Opening of the workshop

Presentation 01 (M.Yamada)

Presentation 02 (K.Ota)

2.2 Session Ia – near-field studies

03: Bentonite analogue study for geological disposal of spent nuclear fuels in Taiwan

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Key words; Zhisin clay, bentonite, buffer/ backfill material, performance assessment, final disposal repository

1. Introduction

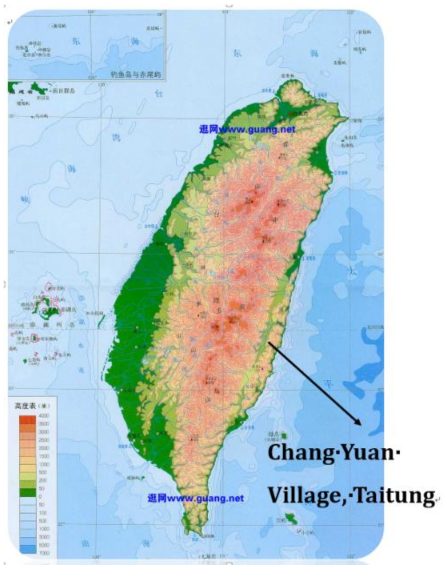
A potential buffer material may be used for radioactive waste repository in Taiwan is a locally available clay material known as Zhisin clay, which is located in Chang Yuan Village, Taitung County, the eastern mountainous mountains of Taiwan The clay has been identified as a Ca-bentonite and the main mineral is montmorillonite. Its formation occurred under the ground from the tertiary volcanic ash, tuff, or andesite agglomerate by the action of hot water and geological changes. The clay in this area has a thick vein shape, which has been developed in the contact zone between the andesite or andesite agglomerate in the Tuluanshan formation and the black shale fault in Takangkou formation. The color of clay is grayish green to white and has good adhesion.

According to the borehole data, approximately 200,000 tons could be mined and the clay reserves at a depth of about 20 meters are quite abundant. The topsoil is a combination of acidic and swelling clay, and contains part of pyrite (FeS_2) which is easily to be oxidized to generate corrosive sulfuric acid and is considered to be less suitable for use in disposal sites. However, it is the only swelling clay rich in montmorillonite in Taiwan. The amount is not high for commercial application and only is mined on the surface. If deeper mining is possible, the content of pyrite can be greatly reduced, and it becomes a potential buffer/ backfill material. The clay sample used in this study was subjected to initial beneficiation. After being crushed and sieved, the sample clay was carried out for experiment by passing through a 200 mesh sieve (about 0.074 mm particles).

This paper presents site description, process flow (screening) and an overview of characterization tests conducted on Zhisin clay, including physical (mechanical) property, chemical analysis, cation exchange capacity, X-ray diffraction, thermogravimetry analysis (TGA). The results of performance assessment, batch and diffusion test results for radionuclides also discussed in comparison with other bentonite.

2. Local bentonite source location, outcrop of mining area and processing flow of Zhisin clay

Figure 1 (a), (b) and (c) show the local bentonite production site in geological map of Taiwan, the outcrop and clay appearance, respectively. Figure 1 (d) shows the beneficiation flow of Zhisin clay, including mining, stacking, crushing, storage, screening, drying, milling and bagging-



b

a) c)

d)

Figure1 (a) The geological map of local clay material; (b) outcrop of mining area (c) appearance and (d) processing flow of Zhisin clay

3 Characterization results

Table 1 displays chemical compositions from XRF results. The iron content (Fe_2O_3) of Zhisin clay is higher than that of MX-80. Iron oxides may be coated on the surface of clay minerals or mineral phases (such as pyrite) are independently formed in the clay, which become the exchangeable cations and exchange with existing Ca^{2+}, Mg^{2+}, Na^{+}, K^{+} and other cations. As listed in Table 2, the CEC (Cation Exchange Capacity) was measured by Inductively Coupled Plasma (ICP) analysis. Zhisin clay is Ca-bentonite, whereas MX-80 is Na-bentonite.

Table 1 Chemical compositions of MX-80 and Zhisin clay

Compositions (wt. %)	MX-80	Zhisin clay
SiO_2	55~64	55.4~57.6
Al_2O_3	18~21	16.9~23.2
Fe_2O_3	2.5~2.8	5.9~10.2
CaO	0.1~1.0	1.6~3.9
Na_2O	2.5~2.7	0.6~0.9
MgO	2.5~6.2	1.5~2.0
K_2O	0.2~0.4	0.3~1.2
SO_3	---	2.5

Table 2 CEC of MX-80 and Zhisin clay

	MX-80	Zhisin clay
CEC (meq/100g)	79	70
K^+ (meq/100g)	2.3	1.4
Na^+(meq/100g)	56	23.1
Ca^{2+}(meq/100g)	30.1	38.7
Mg^{2+}(meq/100g)	15.6	6.8
Na^+/Ca^{2+} ratio	1.86	0.6

Figure 2 (a) shows that Zhisin clay contains the mineral phases of montmorillonite, kaolinite, pyrite, quartz, illite, calite and gypsum, whereas montmorillonite and quartz are dominate in MX-80. The diffraction peak of pyrite can be identified in Zhisin clay, which is consistent with higher Fe and S contents in the chemical analysis. The intensity of the main peak, quartz in the Zhisin clay (2θ = 26.67°) is relatively higher than that of MX-80, indicating that the non-swelling mineral content in Zhisin clay is higher than that of MX-80. As shown in figure 2 (b), the ratio of sandy content (> 74 μm), silt content (74 μm ~2 μm) and clay content (< 2 μm) in the different buffer materials can be estimated and summarized in Table 3.

(a) (b)

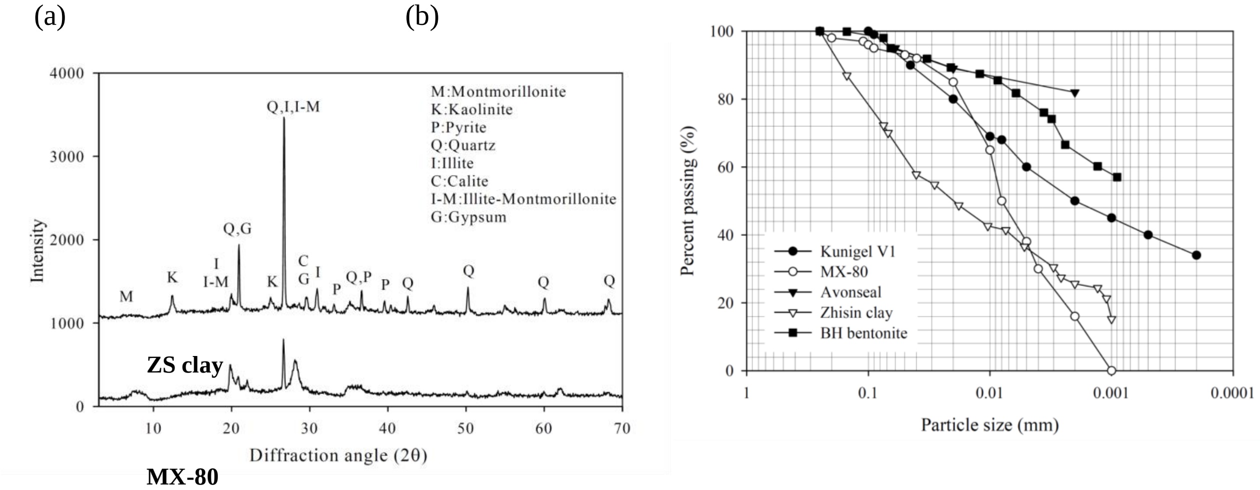


Figure 2 (a) X-ray diffraction patterns of Zhisin (ZS) clay and MX-80; (b) Particle size distribution of each type of bentonite

As can be observed in Table 3, the sandy content (non-swelling part), 28% in the Zhisin clay is much higher than that of MX-80. From the basic characteristics of soil mechanics in this table, it can be found that the property of the Zhisin clay is far from MX-80, which may be resulted from the different types of exchangeable cations, i.e., the former one is calcium bentonite, and the latter one is sodium bentonite.

Figure 3 shows TGA analysis of Zhisin clay and BH bentonite. The latter one, BH bentonite has a two-stage significant weight loss process. The first stage is a mass change and rapid weight loss during the heating process from room temperature to 140 ° C. This stage should be caused by the removal of adsorbed water on the surface of both of clay, indicating dehydration reaction. The second weight loss process occurs between 600 and 700 ° C and is resulted from the removal of structural water (hydroxyl) from the BH bentonite, demonstrating de-hydroxylation reaction. The TGA curve of the Zhisin clay presents a three-stage weight loss process. Compared to BH bentonite, the second dehydrogenation reaction occurs between 450 and 550 °C. Since this stage of weight loss is a critical temperature at which the clay maintains its structural integrity. Therefore, the thermal stability of BH bentonite is better than that of Zhisin clay. The third stage of weight loss process of Zhisin clay occurs between 750 and 850 °C, suggesting that the impurity, $CaCO_3$ may be decomposed in this clay.

Table 3 Physical properties MX-80 and Zhisin clay

Property		MX-80	Zhisin clay
Specific Gravity		2.7	2.67
sandy content (> 74 μm)		5	28
silt content (74 μm ~2 μm)		79	46
clay content (< 2 μm)		16	26
Atterberg limits	Plastic Limit, PL	70	18
	Liquid Limit, LL	400	67
	Plastic Index, PI	330	49
	Activity, AC	4.02	1.88

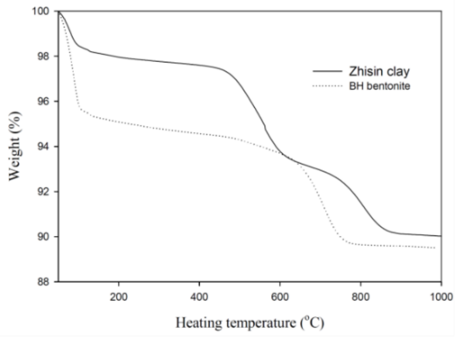


Figure 3 TGA analysis of Zhisin clay and BH bentonite

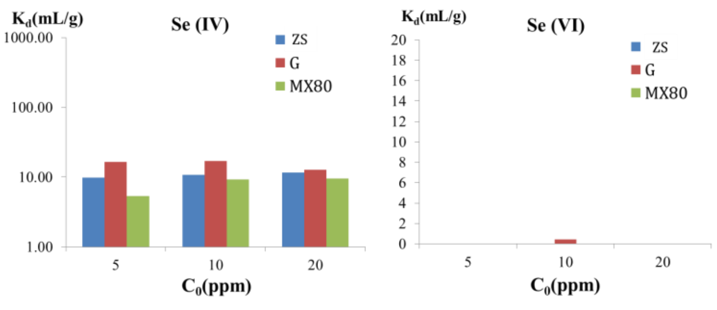
Table 4 lists Cs, Sr and Co sorption on MX-80 and Zhisin clay. The sorption rate (99%) of Sr on MX-80 is much higher than that of Zhisin clay (84%) as the latter one is Ca-type bentonite and may compete with divalent strontium (Sr).

Table 4 Cs, Sr and Co sorption on MX-80 and Zhisin clay

Element	MX-80		Zhisin clay		Liquid Phase
	Sorption (%)	Kd value	Sorption (%)	Kd value	
Cs	94	470	96	725	DIW (Deionized Water)
Sr	99	2970	84	163	
Co	99	2970	99	2970	
Cs	88	228	82	137	SGW (Synthetic Ground Water)
Sr	59	44	42	22	
Co	95	608	95	548	

As observed in figure 4, the Kd values of Se (IV; SeO_3^{2-}, selenite) show higher uptake capacities than those of Se (VI; SeO_4^{2-}, selenate) sorption on Zhisin (ZS) clay, granite (G) and MX-80, as for Se (IV), the FeOSe(O)O(-) inner-sphere complex may be formed onto the hematite (Fe_2O_3) surface. Furthermore, the iron oxide, Fe_2O_3 of Zhisin clay is higher than that of MC-80 in Table 1, indicating higher Kd values for Se (IV) sorption on Zhisin clay under three different initial concentration of (IV).

Figure 5 shows the I-129 diffusion curve in Zhisin clay using through-diffusion technique and the apparent diffusion coefficient (Da) can be obtained between 1.51 to 2.36×10^{-12} m^2/s according to the different kinds of porosities and density. The Da values decreased with an increase in the bulk dry density of the compacted bentonite.



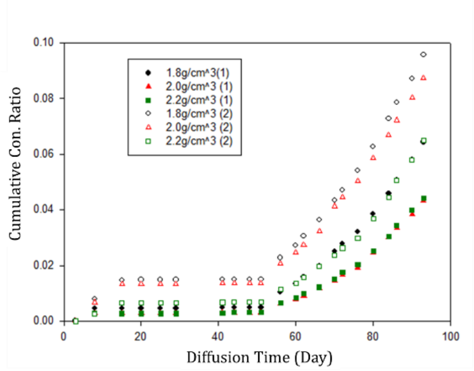


Figure 4 Se (IV) and Se(VI) sorption on MX-80 and Zhisin (ZS) clay

Figure 5 I-129 diffusion curve in Zhisin clay

4. Conclusion

Although Zhsin clay has good adsorption capacity for radionuclides, the chemical and mechanical properties as well as thermal stability are not well applicable to the important criterion of engineering barrier if it is used as a buffer or backfill material for high-level radioactive waste of final disposal repository. In addition, the original mining site has been abandoned for more than 20 years due to less commercial application and without economic values, which results in cost in-effectiveness for mining. However, it can be considered as an evidence of natural analogue study for local bentonite.

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2.3 Session Ib - Mini-session on archaeological analogue studies

Czech Programme of Archaeological Analogues for Modelling of Long-Term Lifetime of DGR Canister

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Key words: archaeological analogues, carbon steel, corrosion products, modelling

Extended Abstract

Czech DGR (deep geological repository) programme focused on canister corrosion is running since 2009. There were three concepts at the beginning: carbon steel/copper, carbon steel/titanium and stainless steel/carbon steel. During the previous projects was the main attention focused on stainless steel/carbon steel concept, because uncertainties of corrosion attack on copper caused by microbial activity and pore solution radiolysis, and possibility of titanium hydrogen embrittlement. Short-term studies in anaerobic conditions of saturated compacted bentonite were done and modelled, showing the lifetime of the canister in order 10^5 years. Nevertheless, the extrapolation to such long-term period based on short-term exposures up to 6 years does not seem serious. Thus, the project of collecting long term data from archaeological analogues was started.

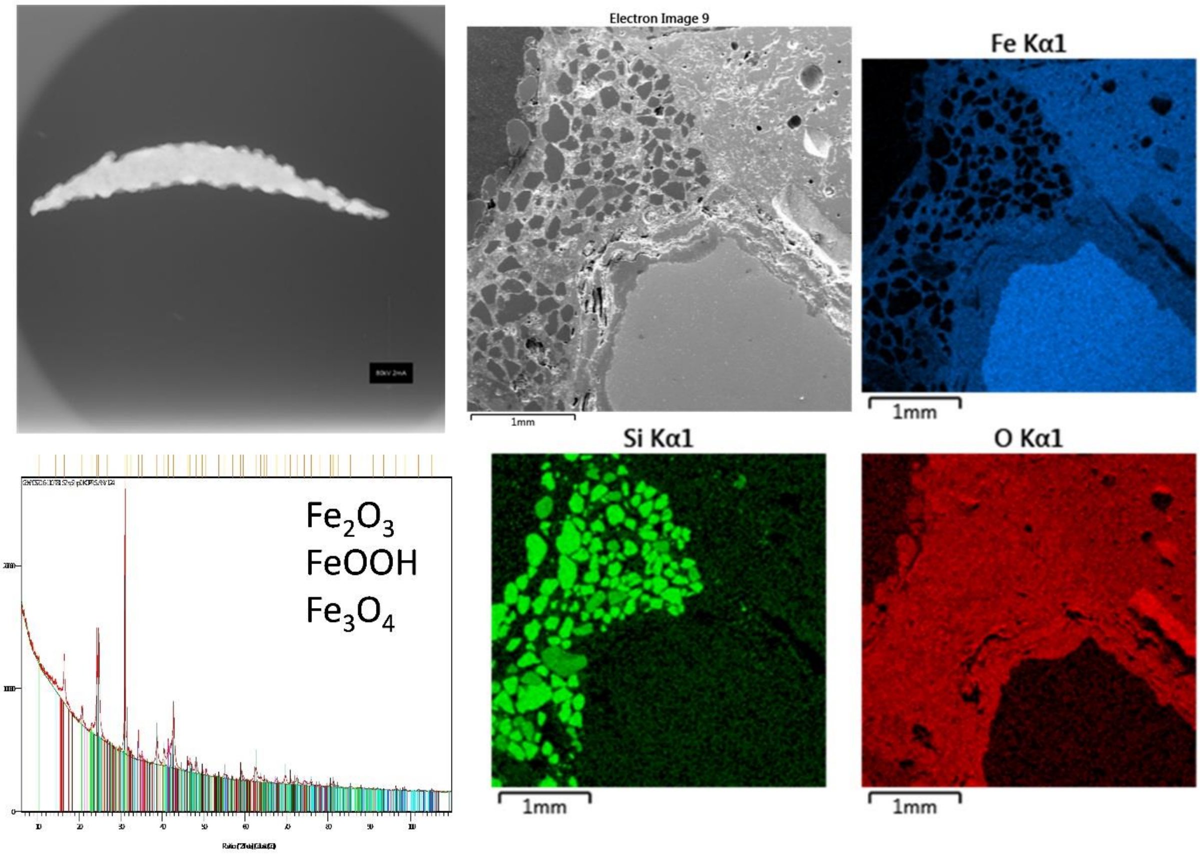
The main focus is given to localities with similar environment as is expected in DGR, i.e. clay bedrock and permanent moisturising. Ideal sites are pond beds or river/creek deltas. When preliminary detector prospection is successful, the locality is subjected to excavation. All collected artefacts are immediately dried and the X-ray imaging is done. A depth of excavation and artefact dating are recorded. Some low value artefacts are subjected to destructive analyses. They are cut and embedded into epoxy resin, polished and observed by electron microscopy (SEM) and elemental mapping (EDS). Corrosion products are ground and analysed by X-ray diffraction.

Soil analyses proceeds as well. There are estimated basic parameters: humidity, dry density and particle mesh analysis. Soil pore solutions are sampled immediately during excavations or there are carried out laboratory experiments with high pressure squeezed pore solution or soil liquors analyses. Lysimeters were installed at suitable localities in order to collect the soil pore solution continually within all seasons.

In general, all soil particles are much larger compared to bentonite particles. Composition of soil pore solutions differs significantly between the localities, but there is no obvious effect on corrosion products composition or corrosion rate. The corrosion rate is estimated based on corrosion products thickness and split to the anaerobic and aerobic contribution according to the corrosion products composition. The aerobic corrosion rate is compared to the model of oxygen transport into the soil in the locality and depth of the artefact.

An important observation was achieved on samples by means of SEM/EDS mapping. Corrosion products have significant crystallisation pressures and they can displace soil particles. Corrosion products are mechanically disrupted in cycles and the layer of corrosion products is formed epitaxially at the metal/corrosion products interface, unlike the mechanism of formation observed during early stages of exposure in laboratory experiments.

Graphical Abstract



Archaeological analogue studies at JAEA

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Key words; natural analogue, archaeological analogue, carbon steel, copper, iron, bronze, overpack, corrosion

1. Introduction

Carbon steel, copper and titanium are considered as candidate materials for a metallic container (overpack) for the geological disposal of high-level radioactive wastes in Japan. Among these candidate materials, carbon steel and copper are commonly found as either iron or bronze artifacts at archaeological sites, and so have been used as “archaeological analogues” to understand the long-term corrosion behavior of overpack materials under geological disposal conditions. This presentation summarizes some recent archaeological analogue studies conducted at JAEA.

2. Iron Artifacts

Ancient Iron Sword

An ancient iron sword (ca. 2,000 years old), excavated from Ohtake-nishi archaeological site in Osaka Prefecture, is one of the oldest cast iron swords in Japan. Because of the good state of preservation, such as the sharply-defined "Shinogi" (the longitudinal ridge) on the surface, the relationship between the burial environment and the corrosion state was studied (Mitsui et al., 2012). At the site, environmental conditions such as the oxygen level, Eh, pH were investigated, and *in-situ* corrosion rates of iron were estimated with a corrosion rate monitor with a carbon steel probe (CRM). The corrosion state of the iron sword was analyzed by corrosion depths using an X-ray computed tomography (CT) and corrosion products using a portable X-ray diffractometer equipped with X-ray fluorescence spectrometer (XRDF). It was found that the redox potential and dissolved oxygen level were very low, and that the long-term corrosion rate of 7.5×10^{-4} mm/y determined from CT-measured corrosion depths, was two orders of magnitude lower than the *in-situ* corrosion rate of 3.2–5.2×10^{-2} mm/y determined by CRM. Covering the iron sword was a 0.4–1.0 mm thick layer composed mainly of siderite ($FeCO_3$), which had formed as the corrosion product.

Ancient Socketed Iron Axe

A well-preserved ancient socketed iron axe (ca. 1,400–1,700 years old) was excavated from Yoshida Nakamichi site in Tottori City, Tottori Prefecture. To understand the reasons of the good state of the iron axe, the relationship between the burial environment and the corrosion state was studied (Mitsui, 2015). Groundwater chemistry, and corrosion rates and products of the iron axe were similarly investigated using CRM, X-ray CT and XRDF, etc. It was found that the redox potential and dissolved oxygen level were again very low, and that the long-term corrosion rate of 0.2–5×10^{-3} mm/y determined from the CT-measured corrosion depths was one to two orders of magnitude smaller than the *in-situ* corrosion rate of 5×10^{-2} mm/y determined by CRM. Again, a 0.4–6.4 mm thick layer of siderite had formed as the main corrosion product.

The good preservation state of the iron sword and axe and the difference between the *in-situ* and the long-term corrosion rates were thought to be a consequence of the siderite corrosion product acting as a protective layer. This common feature of iron corrosion provides supporting evidence for a predictive long-term iron corrosion model described by a power-law equation derived from 10-year iron corrosion tests in compacted bentonite under anaerobic conditions by Taniguchi et al. (2010), where siderite, as well as other carbonate compounds, was identified as the main corrosion product.

3. Bronze Artifacts

Ancient Bronze Bells and Bronze Halberds

Well-preserved bronze artifacts comprising five bells and eight halberds (ca. 2,000 years old) from the Yayoi Period were excavated at the Yanagisawa archaeological site in Nakano City, Nagano Prefecture. Comprehensive analysis of soil and groundwater samples at the site was carried out and geochemical calculations were made to better understand the local conditions that led to these bronze artifacts being so well preserved (Mitsui et al., 2018). Analysis of the soil surface adjacent to the bronze artifacts identified cuprite (Cu_2O) and cassiterite (SnO_2) as the corrosion products. Migration behavior of the bronze metal components, copper, tin, and lead, both inside and outside of the burial pit, was also investigated. Copper and lead had migrated 2 m from the burial pit, whereas tin was confined to the immediate vicinity of the bronze artifacts. The difference in migration behavior of these elements can be explained in terms of the chemical stability of the solid phases. Robbiola et al. (1998) proposed that a tin-enriched corrosion product can act as a protective layer which preserves the original surface of bronze artifacts. This implies that the main factor contributing to the well-preserved state of the bronze artifacts was the tin content, which had formed a protective layer of cassiterite on the outer surface of the bronze artifacts.

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2.3 – Session 1c Mini-session on PNAP – low alkali fluid reaction with bentonite

The Philippines International Analogue Project: An Overview of the Project

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Key words; Natural Analogue, Bentonite, Hyperalkaline Plume, Alkaline Alteration Process, Philippines

1. Introduction

An hyperalkaline environment, high pH, high Ca content, reducing and 30-40˚C, formed by serpentinization of ultramafic rocks in ophiolite can be considered a natural analogue of a TRU waste repository in Japan where significant amounts of cementitious materials will be used.

Many serpentinizing ophiolites are distributed in the Philippines as shown in Fig. 1. The Philippines International Analogue Project have started in 2007 at the Saile bentonite mine of the eastern end of Zambales ophiolite, northwest of Luzon. In this paper, it is shown that an overview of the project implemented mainly at the Saile mine in Luzon and Narra on Palawan as natural analogue of the interaction between bentonite and hyperalkaline plumes.

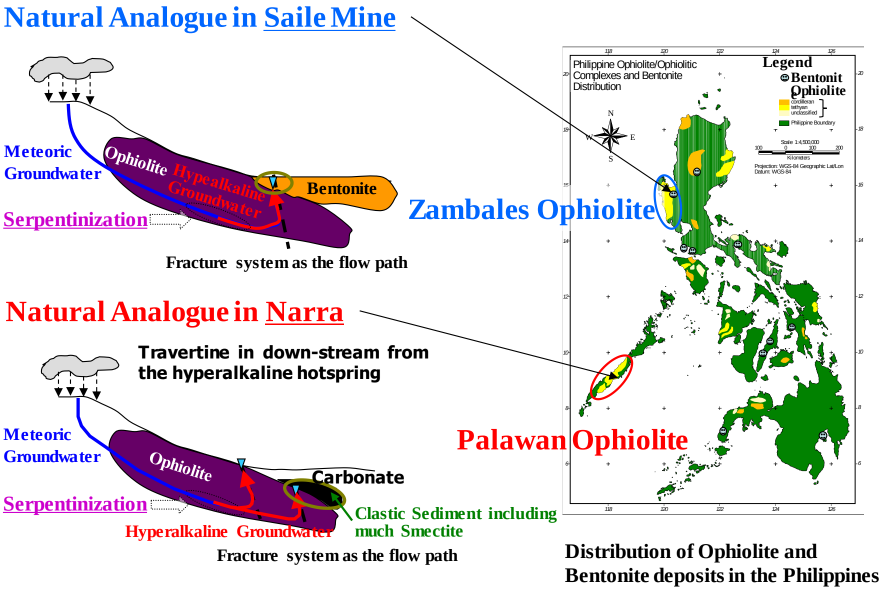

Figure1. Geological Setting in the Philippines

2. Natural analogue site

All hyperalkaline groundwaters studied so far in the Philippines originate from the 20 or so known ophiolite bodies, which are widely scattered throughout the archipelago. In low temperature serpentinisation, meteoric groundwaters react with the ultramafic rocks of the ophiolite in an essentially open system and produce $Ca(OH)_2$-type groundwaters. The hyperalkaline waters in the Philippines are reducing and hot (30-40°C) as shown in Tab. 1. The geochemical properties of these groundwaters, therefore, can be regarded as an analogue of (especially low alkali) cement leachate.

Table1 Groundwater Chemistries in the Philippines

Site	Palawan Narra-1 Hot Spring	Palawan Narra-3.2 Trench 2	Palawan Narra-3.2 DH04	Palawan Narra-3.2 Trench 3	Palawan Narra-3.2 Trench 6	Palawan Narra-3.2 Trench 7	Palawan Narra-3.1 Hot Spring	Luzon Poonbato	Luzon Manleluag Hot Spring - M1	Luzon Bigbiga Well-1	Low alkali cement leachates* (Iriya, 1997)
pH	10.50	11.39	11.39	11.37	10.96	11.16	11.37	11.41	10.80	9.52	11.09
ORP [mV]	-435	-160	-119	-111	-107	-141	-867	-111	-420	8	-
DO [mg/L]	1.94	7.90	2.39	5.70	7.02	3.83	1.19	-	-	-	-
Temp [℃]	47.2	28.8	33.0	33.7	27.2	27.7	38.5	28.0	32.9	29.2	60
CH_4 [ppm]	>700	0	0	0	-	-	200	> 5000	2090	0~560	-
H_2 [ppm]	0	0	0	0	-	-	0	50~1320	0~62	0~130	-
Na^+ [ppm]	96.4	46.1	51.8	50.4	50.1	49.1	50.4	24.3	26.3	100.6	43
K^+ [ppm]	1.46	2.32	2.96	2.39	2.18	2.02	2.36	1.45	0.375	1.05	13
Ca^{2+} [ppm]	3.78	26.9	22.4	48.5	24.6	26.8	50.6	92.5	29.9	1.63	16.8
Mg^{2+} [ppm]	0.17	0.02	0.02	0.01	0.01	0	0.01	0.24	0.03	0.02	-
Si^{2+} [ppm]	86.4	2.5	2.9	2.5	5.77	4.27	0.29	11	17	72.3	
Al^{3+} [ppm]	0.19	0.08	0.06	0.08	0.19	0.18	0.13	1.41	1.05	0.97	0.3
$Fe^{(2+,3+)}$ [ppm]	0.06	0.02	0.03	0.05	0.02	0.02	0.01	0.18	< 0.001	0.0058	-
Cl^- [ppm]	56.5	27.3	21.1	27.0	28.5	28.6	28.0	12.9	17.8	4.50	-
SO_4^{2-} [ppm]	6.34	0.13	0.14	0.02	1.92	0.63	0.05	0.05	0.364	48.0	-
HCO_3^-** [ppm]	-	0.67	2.8	0	9.2	8.9	0.5	1.6	30.0	135.6	-

*Mix Proportion [wt%] - Portland Cement: Silica Fume: Fly Ash=40: 20: 40, **HCO_3^- - HCO_3^- + CO_3^{2-} from the alkalinity titration

One of study site is distributed nearly at the quarry of Saile bentonite deposit (Saile Mine) located at the eastern edge of the Zambales ophiolite in Luzon. Manleluag hot springs are located in the vicinity of about 2.7 km southwest away from the Saile mine. These high pH spring water derived from the water-rock interaction with the active serpentinisation and have been gushed and seeped out along the fracture systems. in the Zambales ophiolite.

The bentonite bed of Aksitero formation is overlaid directly on the pillow basalt of Zambales ophiolite. The main constitute minerals of bentonite in the Saile mine is composed of Ca-montmorillonite and accessory minerals are Ca-zeolites, calcite, plagioclase, rarely a few of clinopyroxene, opaque minerals and silica minerals.

Another study site is Narra located in central Palawan. Here, hyperalkaline groundwater are currently seeping into overlying clastic sediment. Geochemical properties of hyperalkaline groundwaters are almost same as that of Manleluag in Luzon.

The upper layer at the study site is formed with carbonate from surface travertine. The lower layer is formed with the black clastic sediment and overlies the basement ultramafic rock of harzburgite. The clastic sediment is not bentonite but smectite-rich. Reaction time between the clastic sediments and the hyperalkaline groundwaters is about 2,500-9,500 years by 14C dating for humic acid in the clastic sediment by JAEA-AMS-TONO.

3. Result and discussion

Some geological and mineralogical evidences found at Saile mine show that the hyperalkaline groundwater had flowed through fracture systems and had come in contact with bentonite layer. It is observed that bleached zones in bentonite layer near the contact part. The zone is 30-40cm wide from the surface of pillow basalt and consists of Ca-smectite mainly and zeolites as an accessory mineral. Mineral composition of bleached zone is not different from that of non-altered bentonite zone except for the part of heterogeneous calcite.

The evident alkaline alteration on the contact interface is a width of about 5mm and is constituted of K-feldspar, Ca-zeolite and the iron accumulation band. The iron accumulation band consists of Fe-rich smectite like nontronite and Fe-saponite and the density is higher than a non-altered bentonite zone.

The alkaline alteration zone is limited to mm scale as a result of clogging of Fe-rich minerals. Because of these facts, Natural analogues of Saile mine shows the evidences that the long-term stability of bentonite under hyperalkaline condition have been enough maintained.

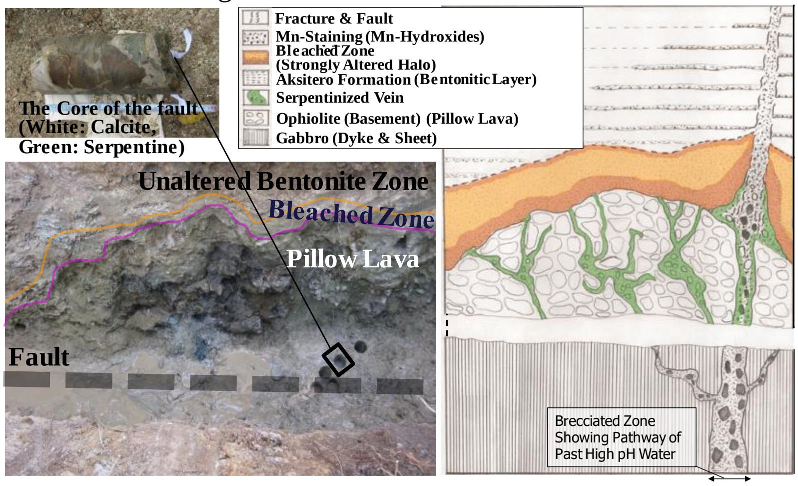

Figure2. Photo and illustration of type section as the past Interaction between bentonite and Hyperalkaline fluids at trench-1 in Saile mine

At Narra site in Palawan, it is possible to observe alkaline alteration processes with dissolution and generation of smectite. Smectites of various types (Fe-Saponite and Nontronite etc.) were identified in clastic sediments underlying the hyperalkaline groundwaters. C-S-H coprecipitated with Fe-Saponite from microscopic observation. It is assumed from all results that primary minerals of clastic sediment are mafic minerals such as olivine and orthopyroxenes, then these minerals reacted with hyperalkaline groundwater and Fe-saponite was precipitated from the hyperalkaline fluids.

Fe, Mg, Si, Al and Ca in the hyperalkaline fluid affected the formation of smectite and the smectite of Fe (Mg)-rich is stable under hyperalkaline condition. That is hyperalkaline fluids with Fe, Mg is preferable for the formation of smectite.

It is the same process at Saile mine that Fe-rich smectite were formed under hyperalkaline condition at Narra. The alkaline alteration process from both of the analogues in the Philippines is summarized in Fig.3. The Philippine natural analogues can show not only the result of long-term stability of bentonite but also the alteration process.

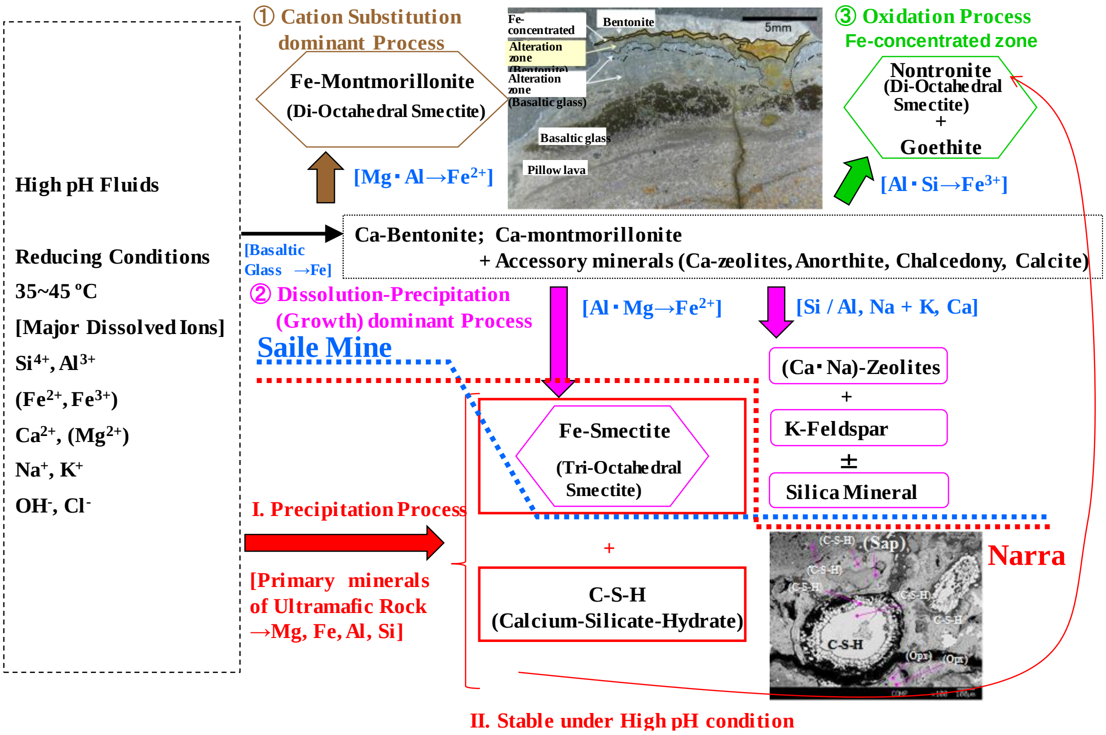

Figure3. Alkaline alteration processes in the Philippine natural analogues

4. Acknowledgement

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Mineralogical evolution of Fe-Mg-Si phases under low-temperature and alkaline conditions at Narra in Palawan, Philippines

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Key words; mineralogical evolution, alkaline condition, Fe-Mg-Si phases

1. Introduction

Bentonite will be planned to use in radioactive waste disposal facilities as an engineered barrier due to its low permeability, which is imparted by its major component, montmorillonite. Cementitious materials are also used as engineered barriers. Alkaline conditions generated by the alteration of cementitious materials may cause the dissolution of montmorillonite, resulting in an increase in the permeability of bentonite. In contrast, the precipitation of secondary products via the simultaneous dissolution of primary minerals may cause a decrease of porosity, resulting in a decrease of permeability. One of the candidates for such a secondary product is an amorphous phase, such as magnesium silicate hydrate (M-S-H) [1]. Such precipitates would be destabilized relatively quickly when chemical conditions change. Even if chemical conditions remained stable for the precipitates, they would transform to stable phases relatively slowly. To evaluate the permeability change of bentonite, it is important to understand the mineralogical evolution of the precipitates.

When examining long-term geochemical reactions which could occur in the disposal facilities, high temperatures need to be used in order to accelerate the reaction in laboratory experiments in a feasible period. The findings from accelerated laboratory reactions can be helpful in understanding geochemical reactions in the disposal facilities. However, the thermal condition used in these experiments is quite different from that at actual disposal facilities. Therefore, understanding long-term geochemical reactions at temperatures prevailing in the disposal facilities is indispensable in addition to that from the laboratory experiments. Understanding natural geochemical processes under alkaline conditions like those caused by the alteration of cementitious materials is an expedient to understand long-term geochemical reactions at acceptable temperatures.

A study on natural long-term geochemical reactions under alkaline conditions was conducted at Narra in Palawan, Philippines. A previous study investigated the mineralogical and geochemical characteristics of collected samples in order to elucidate the formation mechanism of reaction products [2]. However, a limited number of samples was analyzed, and it was uncertain whether the reaction in question was occurring throughout the site. In the present study, spatial distribution of the secondary minerals was investigated to understand the reaction throughout the site.

2. Site description and samples

The geology of the site is dominated by Palawan ophiolite. The site in central Palawan is an alluvial fan channel spreading on a gentle slope of the Palawan ophiolite basement. The basement is composed of serpentinized harzburgite and a small amount of gabbro [2]. Eight trenches (T1 ~ T8) and four drill holes (DH1 ~ DH4) were excavated targeting deposits of clastic sediments originating from Palawan ophiolite. Samples from each trench and drill hole were numbered in order from the bottom to the top.

Alkaline groundwater was found to be seeping into clastic sediments composed of ultramafic rocks. As the alkaline groundwater originated from the interaction between ultramafic rocks and meteoric water [2], alkaline groundwater might have been seeping into the site over a geological time-scale. Chemistry of the alkaline groundwater is as follows; pH >11, ORP of -176 to -30 mV, a temperature of 27–36 °C, and Ca-OH type [2]. These chemical characteristics were similar to those of alkaline leachates from low-alkaline cement.

3. Results

XRD patterns showed that the targeted clastic sediments were mainly composed of crystalline minerals such as serpentine, pyroxenes, amphiboles, and calcite. Smectite and 14Å tobermorite were also identified in some samples. SEM-EDS analysis revealed that infillings between the primary minerals of the sediments could be divided into two categories. One was a solid phase mainly containing Si, Fe, Mg, and Al (Mg-type infillings). The other one was a solid phase containing Si and Ca, which was mainly observed in samples with tobermorite.

A homogeneous distribution for chemical components of the Mg-type infillings was observed within the sample as shown in Figure 1. A comparison among the samples showed the chemical compositions of the Mg-type infillings are different. The range of (Fe+Mg+Al)/Si ratios in the Mg-type infillings were generally between 0.5 and 1.5 (Figure 2). Since the (Fe+Mg+Al)/Si ratios of Fe- and Mg-rich smectite generally show around 0.5 ~ 0.8, it was indicated that smectite and other phases with higher (Fe+Mg+Al)/Si ratios were present as Mg-type infillings at the site. With regard to the changes in the chemical composition of the Mg-type infillings, the (Fe+Mg+Al)/Si ratios decreased with sampling depth except for DH4-8 (Figure 2).

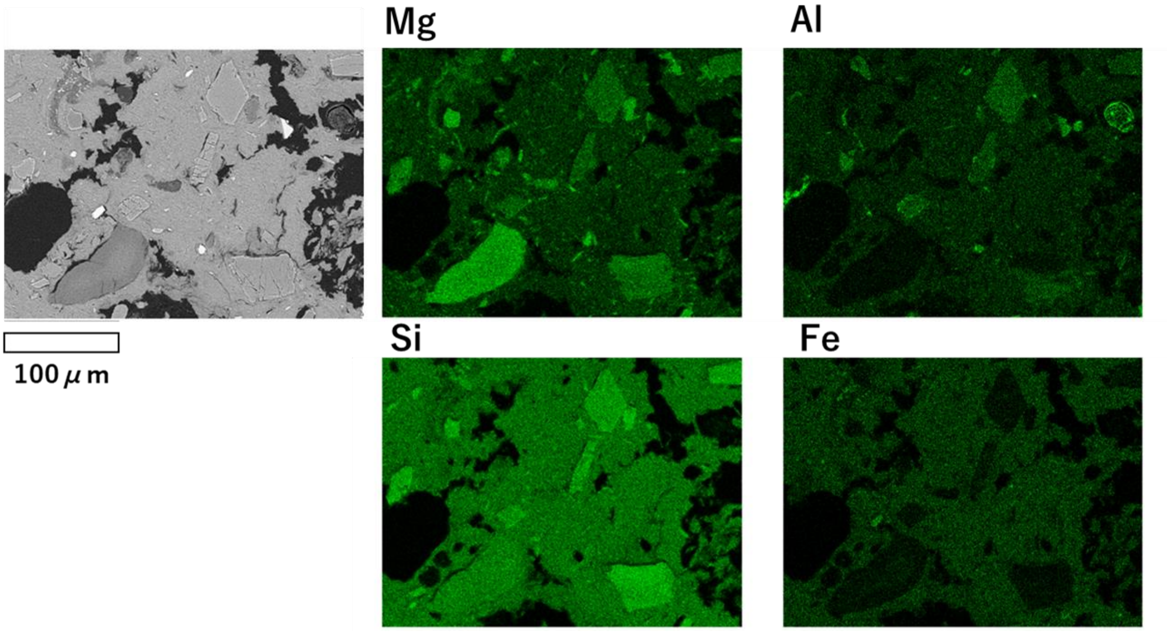

Figure 1. BSE image and elemental mapping of sample T3-4

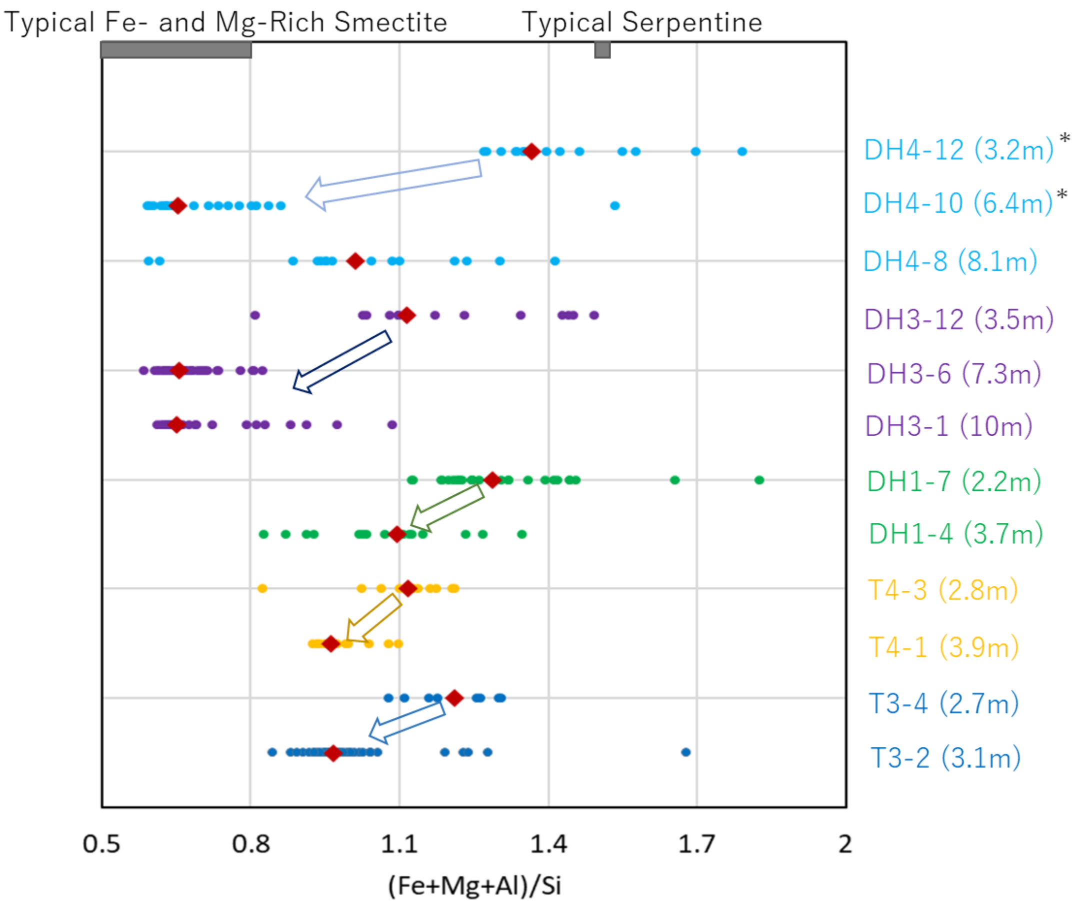

Figure 2. (Fe+Mg+Al)/Si ratios of infillings mainly composed of Si, Fe, Mg and Al.

The diamond plots show the median for each sample. The number in parentheses show depth.
*: Reported in Shimbashi et al. (2018)

4. Discussion

A previous study [2] revealed that the Mg-type infillings with a low Si content in DH4-12 were similar to M-S-H [3] and Fe^{2+}-Si coprecipitate [4]. Furthermore, they indicated that M-S-H were thought to be precipitated from alkaline groundwater at the site and might transform to smectite found in DH4-10 [2]. The present study revealed that decreasing of (Fe+Mg+Al)/Si ratios for the Mg-type infillings with depth was observed in not only the above samples but also samples collected from DH1, DH3, T3, and T4 (Figure 2). On the other hand, chemical composition of the Mg-type infillings showed homogeneous distribution within the samples (Figure 1). Therefore, these findings implied the following: At shallower depths, Mg-type infillings with a low Si content were precipitated by the alteration of the primary minerals. At deeper depths, Mg-type infillings with a high Si content similar to smectite were formed by the transformation of Mg-type infillings with a low Si content, or were formed by direct precipitation instead of Mg-type infillings with a low Si content. The intermediate (Fe+Mg+Al)/Si ratios between Mg-type infillings with a low Si content and smectite in some samples (e.g. T3 and T4) could be due to the presence of a mix of those two phases, or formation of other Mg-type infillings with a medium Si content. To understand time-scale of the transformation or factors which determine species of Mg-type infillings are future issues. Because the sedimentation age can be analyzed by ^{14}C dating of humin or woods in clastic sediments, the time-scale on which the mineralogical evolution of the Mg-type infillings progressed could be estimated, and will be discussed in a future paper.

5. Conclusion

The formation of Mg-type infillings occurred throughout the site. However, the chemical compositions of the Mg-type infillings were different depending on the location. At shallower depths, Mg-type infillings with a low Si content were thought to be precipitated by the alteration of the primary minerals. At deeper depths, Mg-type infillings with a high Si content might have been formed by the transformation of Mg-type infillings with a low Si content, or formed by direct precipitation instead of Mg-type infillings with a low Si content. By estimating the deposition time of clastic sediments at this site, the time-scale of the geochemical reaction at the site will be estimated in a future study. Long-term geochemical reaction of the mineralogical evolution at low-temperature may be useful for the safety assessment of radioactive waste disposal.

Acknowledgements

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Comparison with the results of the Cyprus NA Project (CNAP)

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Introduction

Bentonite is an important component in many designs for radioactive waste repositories. The plasticity, swelling capacity, colloid filtration, low hydraulic conductivity, high retardation of key radionuclides and stability in relevant geological environments all make bentonite an ideal barrier/buffer material. However, bentonite is unstable in higher pH environments and this is a potential problem for repository designs which mix cement and concrete with bentonite barriers. The alkaline (initial pH~13) leachates from the cement are expected to degrade the bentonite, with it potentially losing some or all of its favourable properties. This has 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 reaction, so allowing the use of low alkali cements in close proximity with bentonite (cf. discussions in Alexander & Neall, 2007).

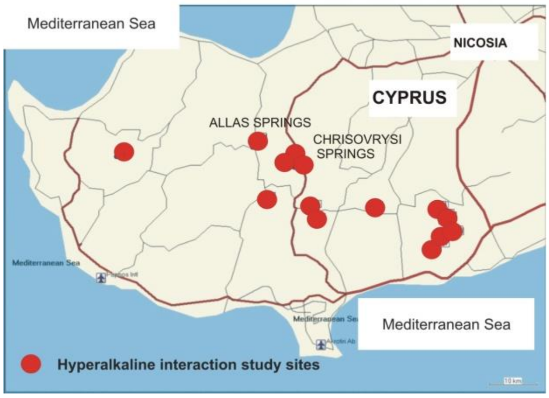
Assessing the long-termed stability of bentonite in contact with such alkaline fluids under conditions representative of a deep geological repository requires complementary laboratory, modelling and *in situ* studies. In particular, to build a robust safety case, it is important to have supporting natural analogue data to confirm understanding of the likely long-term performance of bentonite.

Natural analogue studies could:

- Provide information on reaction 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 reaction
- Provide input to a range of supporting documents for safety cases (cf. Posiva, 2013; Reijonen, 2021))

Although systems representative of leachates from both OPC (i.e. groundwaters with pH 12.5 and above) and low alkali cements (groundwater pH of 9 to 11) have previously been examined as natural analogues of cementitious repositories (e.g. Pitty & Alexander, 2011; Shimbashi et al., 2021, respectively) neither produced data of relevance to the questions now being posed with regard to low alkali cements. For example, the natural cements in Jordan are closely representative of OPC-based materials, so the pH is too high, whereas the work of Shimbashi et al. (2021) did not examine the interaction of alkali solutions with bentonite. An appropriate low alkali water chemistry was also studied in Oman (e.g. Bath et al., 1987), but this project did not include any investigation of bentonite/leachate interaction.

There are a number of locations worldwide where an appropriate natural analogue might be found, including Oman, California, Bosnia, Papua New Guinea and the Philippines. Based on a multi-attribute analysis, considering factors such as probability of finding suitable locations with natural alkaline groundwaters and natural bentonite together, relevance to European national radioactive waste disposal programmes and logistics (including ease of travel and cost-effectiveness), Cyprus was chosen as the focus for this study. Several sites in Cyprus were selected as particularly promising for this purpose; preliminary field investigations in early 2009 confirmed the presence of hyperalkaline springs (pH of 10-12) in the vicinity of bentonite and clay-rich soils (see Figure 1).



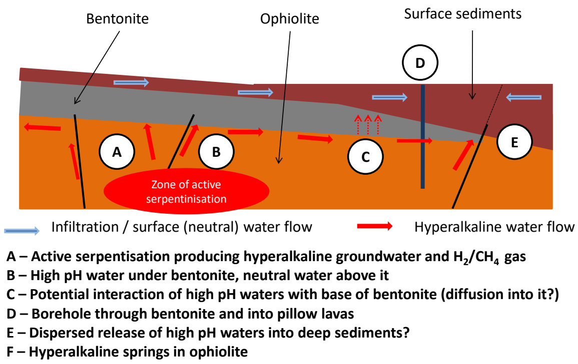


Figure 1: Preliminary investigations have identified over a score of relevant sites in southwest Cyprus (from Milodowski et al., 2009).

Figure 2: The natural system in Cyprus. Alkaline groundwaters could react with bentonite wherever they meet (from Alexander and Milodowski, 2011).

All alkaline groundwaters studied so far in Cyprus originate from ophiolite host rocks which are wide-spread across the island. The alkaline pH values (generally between pH 10 and 11) observed in the groundwaters are a product of the serpentinisation of the ophiolites**.** The presence of bentonite and other clay-rich rocks in close proximity to the natural hyperalkaline groundwaters (Figure 2) permits the zones of potential bentonite/alkaline water reaction to be studied as an analogy of the reaction zones in the repository.

Results

- The results suggest that there has been some reaction (smectite-to-palygorskite transformation – see Figure 3) in the bentonite, probably due to alkaline groundwater interaction
- Both geomorphological and U/Th data rule out early hydrothermal alteration and the low-temperature groundwater system has been operational for 500-800 ka
- Reaction from infiltrating surface waters seems unlikely as the soil and bentonite appear to be very tight

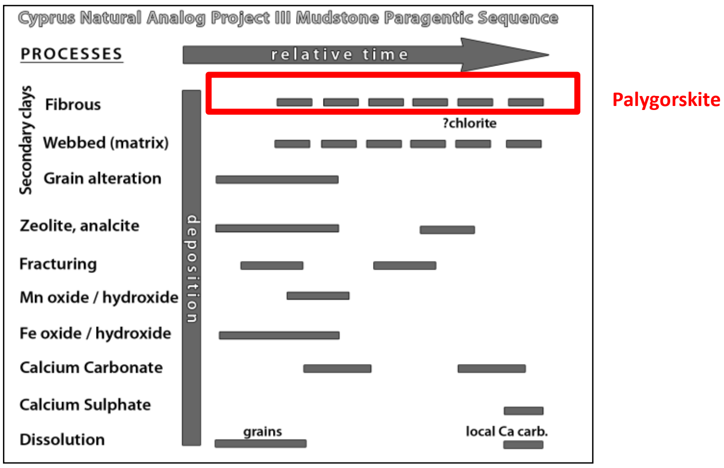


Figure 3: an overview of the secondary phase paragenesis in the samples from the Parsata site in cyprus (Alexander & Milodowski, 2014)

Conclusions

- Reaction of bentonite is restricted to the base of bentonite and to within several millimetres of fractures in the bentonite
- This is probably not due to a limited supply of OH- (cf. 500 – 800 ka of circulation)
- Probably more likely due to very tight bentonite
- It is possible that the smectite-to-palygorskite transformation is a fast reaction (cf. Moyce et al., 2014)
- Palygorskite is a common authigenic clay mineral in “alkaline lake” environments, evaporative soil environments (where alkaline pH is implicated), and from serpentinisation in modern oceanic floor

Sato et al., (2021) claim to have observed secondary iron reactions in the natural bentonites of the Philippines following reaction with alkali groundwaters. However, these results do not address the fact that the sampling zone is less than 1 m deep and that a clear redox front lies immediately above the supposed reaction zone. In addition, no ages for the system in question have been provided, so the temporal scale of the reaction in also completely umknown. As such, until it can be conclusively shown that the surficial nature of the sampling site in this case had no influence on the observed results, it is argued here that long-term reaction of bentonites under the pH conditions expected in the vicinity of low alkali cements in a repository is unlikely.

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T.Sato, M.Shimbashi, N.Fujii, M.Yamakawa, T.Kasama & T.Otake (2021). Bentonite-alkaline fluid interaction at bentonite deposit in Philippines: pore clogging by secondary precipitates formed by the interaction. *These proceedings*.

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Bentonite-alkaline fluid interaction at bentonite deposit in Philippines: pore clogging by secondary precipitates formed by the interaction

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No abstract available.

3. Session II – Poster Session

Application of non-destructive methods in bentonite analogue research

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Key words; X-ray computed tomography, hyperspectral analysis, bentonite, natural analogue

1. Introduction

Samples from various natural clays have been investigated by X-ray computed tomography (XCT) in order to enhance the spatial understanding of the textural variation, and to assess the applicability of the method. Samples from fracture gouges (clays) and bentonite deposits have been examined. Detailed examination of bentonite samples has been focussed on the textural and deformation features and assessing the similarity of natural and engineered systems to reduce uncertainties related geological information to be used to support performance assessments of the bentonite based engineered barrier system materials (EBS). For the fracture clays, the main emphasis has been set to describe different fracturing systems and to asses, whether comparable clays to bentonite based EBS can be observed in the fractured environment similar to that of Finnish geological repository host rock at Olkiluoto. In addition, hyperspectral scanning method has been tested on two sets of samples.

2. Samples and methods

Samples investigated in this study have been collected from Finland, Cyprus and Japan (Figure 1). The samples from Finland represent fracture gouge clays, and have been collected from two localities (sample amounts provided in brackets): Hyrkkölä (3) and Olkiluoto (16). Bentonite samples are from Kato Moni (5), Cyprus and Tsukinuno (3) and Dobuyama (1), Japan. The various types of bentonite samples were used in the testing of the method applicability of XCT (section 3.1). Since this initial phase, the method has been employed in IBL-project (see section 3.2). Samples from fracture clays are part of an ongoing bentonite-rock interaction research project, BROCTIO (see section 3.3).

Figure 1. Locations of the sampling sites; Finland, Cyprus and Japan.

XCT analyses were conducted using a GE phoenix v|tome|x s for all samples. Hyperspectral (2D/surface mapping) scanning was performed on the samples from Cuprus only using SisuChema device. BROCTIO samples from Finland were analysed using hand held Terra Spec HALO. Both hyperspectral analyses were performed to test the applicability of the hyperspectral method on bentonite mineralogical characterization.

3. Preliminary results

3.1 Initial assessment of the method

Initial assessment of the XCT and hyperspectral analysis on natural bentonites was done using existing samples from the Kato Moni field investigations reported by Alexander et al. (2017). Preliminary assessment of the XCT method shows that by 3D imaging the drill core samples using XCT the reliability of textural and deformation analysis could be further improved. Especially deformation related structural features can be investigated without the disturbance caused by e.g. thin section making process providing a view of the sample resembling more *in situ* state. Potential drilling artefacts are also readily visible in XCT models of bentonite, since the density changes due to rotation can be observed (Figure 2).

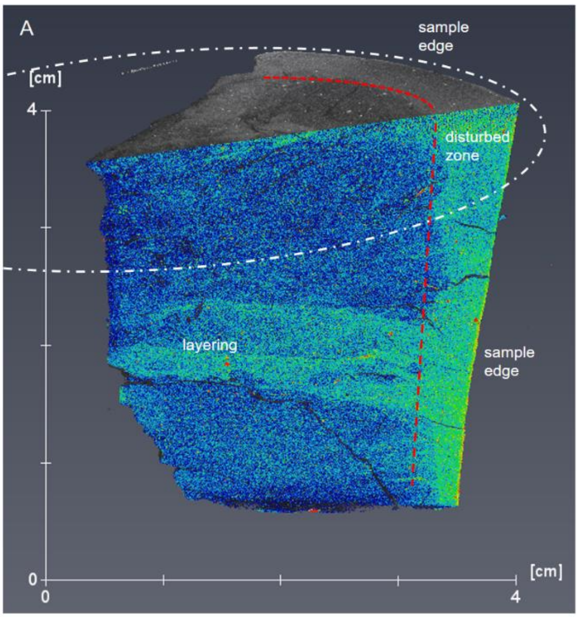

Figure 2. An example of drill core section from Kato Moni bentonite showing clear disturbance at the edge of the sample due to drilling. Original layering also visible in the middle section of the sample (from Reijonen et al. 2020).

Kato Moni samples contain a suite of accessory minerals and microfossil material that provide a good contrast for XCT method, providing clear density differences. Tsukinuno and Dobuyama samples were added to the investigation to broaden the mineralogical variation in the bentonites investigated. Tsukinuno deposit contains high-grade almost pure smectite bentonites, as well as sandy bentonite variants providing a good compositional analogues to various types of EBS bentonites. Dobuyama sample was added due to its apparent pelletal texture, this sample has been further investigated in the IBL project (Reijonen et al. 2019a). To be able to enhance the preliminary analysis, also two manufactured bentonite samples were included in the tests, compacted bentonite block (Wyoming Na-bentonite) and commercial pellet bentonite.

Hyperspectral scanning results from Kato Moni samples has produced more detailed mineralogy than what was observed in the original XRD (from Alexander et al., 2017). Further analyses are planned to confirm the promising preliminary results.

3.2 Application of the method in bentonite analogue study

XCT was applied to Tsukinuno bentonite samples in the IBL project and the results will be reported as part of the full interpretation of the investigations. Some preliminary results have been presented (Reijonen et al., 2019a,b). XCT imaging was successfully performed on drill core samples to obtain full 3D model of the bentonite sampled, as well as on the hand specimens (Figure 3).

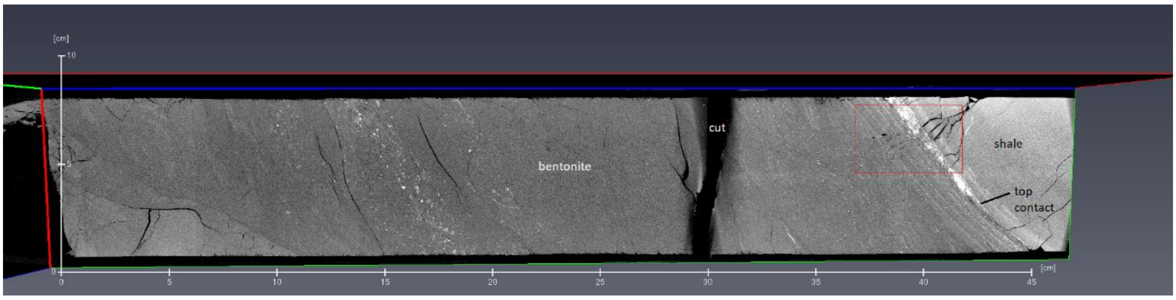

Figure 3. An example of XCT cross section of the bentonite drill core sample (Reijonen et al. 2019b).

3.3 Application of the method in facture mineral study

Based on the good experiences in the bentonite studies discussed above as well as those reported in the literature (e.g. Kuva et al., 2012) XCT was chosen to be applied also for fracture gouge samples in BROCTIO-project. Within the project also bentonite samples will be studied in connection to experimental analysis. XCT is supported by additional examinations, including SEM, XRF and XRD. In addition, a hand-held hyperspectral measurements have been performed and planned to be verified using XRD. Preliminary results show that the fracture systems can be observed via XCT, however, clay phase identification especially needs supporting methods. The properties that can be observed include: textural properties of fracture fillings, texture of the unaltered rock, open fractures in samples, density differences in fracture fillings (Figure 4).

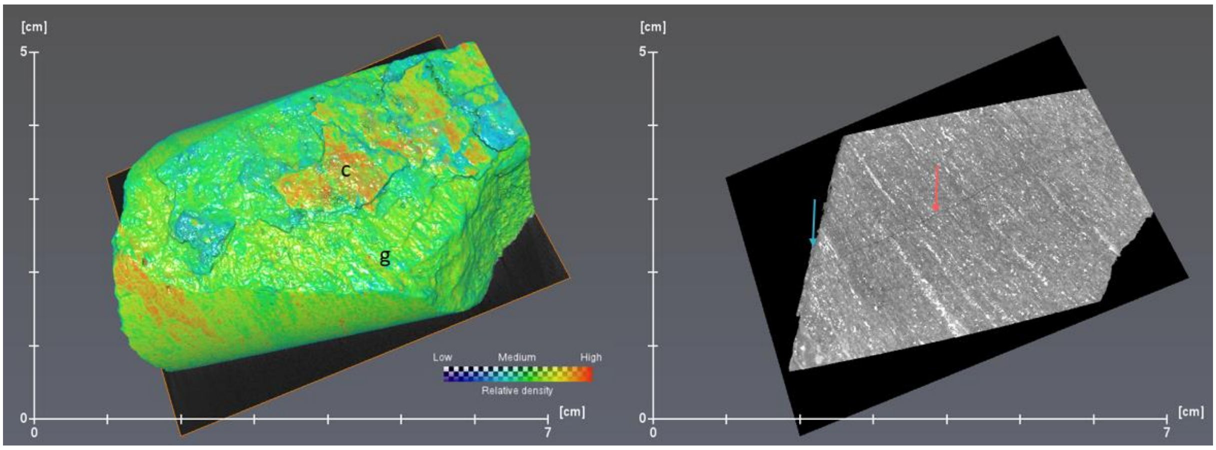

Figure 4. 3D image of the granitic sample (from Hyrkkölä) with two clay bearing fracture surfaces. Left: Clay layer (c) on top of the granitic sample in fracture surface (g). Right: Internal open fracture in the granitic sample (red arrow) and cross section of the fracture filling clay (blue arrow).

4. Summary

Application of non-destructive methods on bentonite analogue research has proven to be informative and significantly adding to the overall analytical scheme to be applied. XCT provides an efficient way to describe clay samples to better plan the subsequent sampling and to estimate sampling related deformation to samples. It is also powerful method to observe textural patterns in natural bentonites. Application to both natural bentonites and bentonites used in experimental set-ups, provides a good measure to assess the physical similarities in the systems. Hyperspectral scanning is still at development stage, but has shown promising results that could be beneficial to obtain first hand data in the field, or to support laboratory analysis (potential use in extrapolation of .g. XRD results).

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Precipitation of magnesium silicate hydrate at ambient temperature under natural alkaline conditions

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Key words; Magnesium silicate hydrate, Alkaline condition, Serpentine, Mineral-water interaction, Carbonate

1. Introduction

Understanding formation of Mg-silicate minerals at ambient temperature is essentially required in various fields of engineering geology such as radioactive waste disposal. The geological disposal of radioactive waste requires semipermanent isolation with an engineered barrier made up of bentonite and cement in tentative Japanese concept. However, in case groundwater flows into the cement barrier and alkaline pore fluids are produced, the fluids might alter bentonite. Previous studies reported that the results from laboratory experiments indicated formation of some magnesium silicate hydrate(s) (M-S-H) as secondary phase(s) around interfaces between cement and bentonite, the fluids supplying Mg and Si from bentonite and cement barriers (Ramirez et al., 2002; Mäder et al., 2017).

However, so far, the most studies were previously discussed only from experimental points of view. Natural M-S-H formation should also be understood because geological conditions in the radioactive waste disposal presumably differ site by site.

In this context, this study conducted investigation of natural precipitates to understand formation of M-S-H under natural alkaline environment.

2. Site description

In order to observe in-situ precipitation of M-S-H under natural alkaline environment, this research focused on the geological setting in the presence of both groundwater and mafic rocks.

In particular, we focused on serpentinite because alkaline seepage would be expected. Serpentinite is distributed

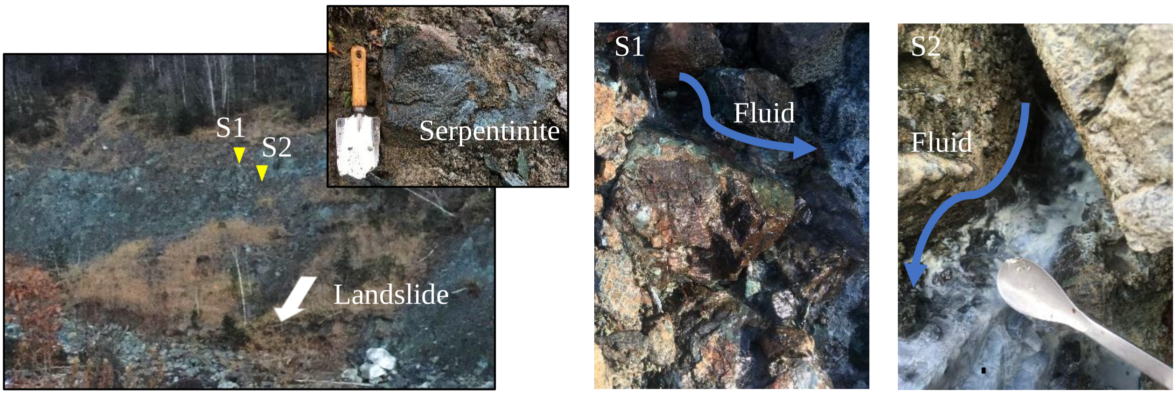

Figure 1. Photographs of sampling sites in the Kamuikotan tectonic belt, Hokkaido, Japan

in various regions around the Kamuikotan tectonic belt in Hokkaido, Japan. Therefore, the samples were collected from two seepages (S1 and S2) on a serpentinite rock mass within the Kamuikotan tectonic belt (Figure 1). Collected effluent samples were the groundwaters from the seepages, and solid samples were grayish/whitish sediments precipitated on the surface around S1 and S2.

3. Results

3.1. Characterization of sediments

X-ray diffraction (XRD) profiles displayed bulk mineralogy of sediments from S1 and S2. Although the XRD profiles of both sediments showed peaks of serpentine which was likely derived from bedrock, peaks of secondary Mg-silicate mineral (i.e., M-S-H) were indiscernible.

On the other hand, in addition to detrital serpentine from bedrock, transmission electron microscopy (TEM) discovered precipitation of M-S-H which was mineralogically identified as low-crystalline chrysotile mainly composed of Mg, Si, and O (Figure 2). The M-S-H was unambiguously different from chrysotile composed of the serpentinite with respect to particle size and crystallinity. Microscope observation of the sediment showed infillings of the M-S-H around detrital particles, therefore, this product has been precipitated from the effluent at ambient temperature.

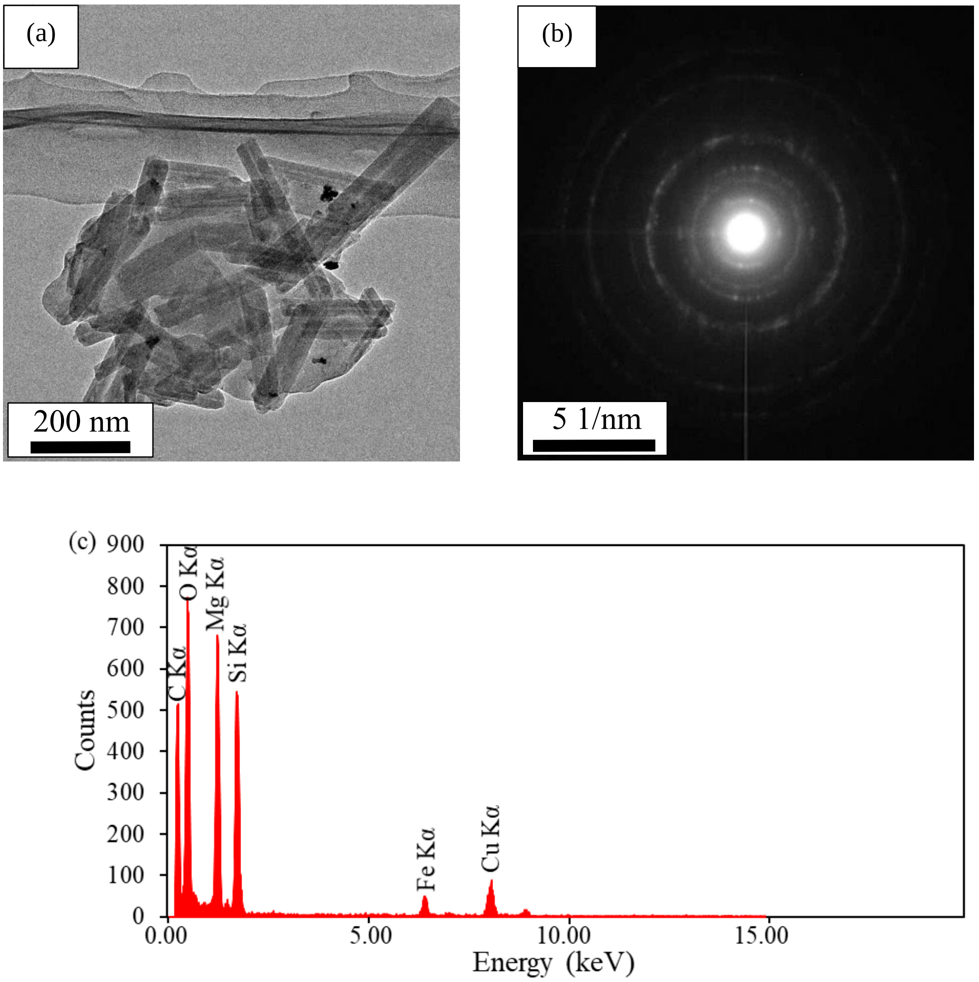

Figure 2. TEM results of some particles in sediments from S1: (a) A TEM image; (b) An electron diffraction pattern; (c) An EDS spectrum of the particles

In addition to M-S-H, XRD and microscopy of the sediment from S2 also showed aragonite precipitation.

3.2. Effluent chemistry and thermodynamics

Collected effluent samples from the seepages showed high pHs (≈ 10) and high alkalinities. Concentrations of dissolved Si were extremely low. Calculating activities of dissolved species with geochemical modelling software brought the results that both samples were saturated with one of M-S-H ($M_{1.5}SH_{2.5}$), which is reported as a low-crystalline serpentine-like material (Nied et al., 2016; Lothenbach et al., 2019) (Figure 3a). Although both samples were supersaturated with aragonite, aragonite was observed only at S2 because the sample from S2 was supersaturated with monohydrocalcite, which was a precursor for aragonite (Figure 3b).

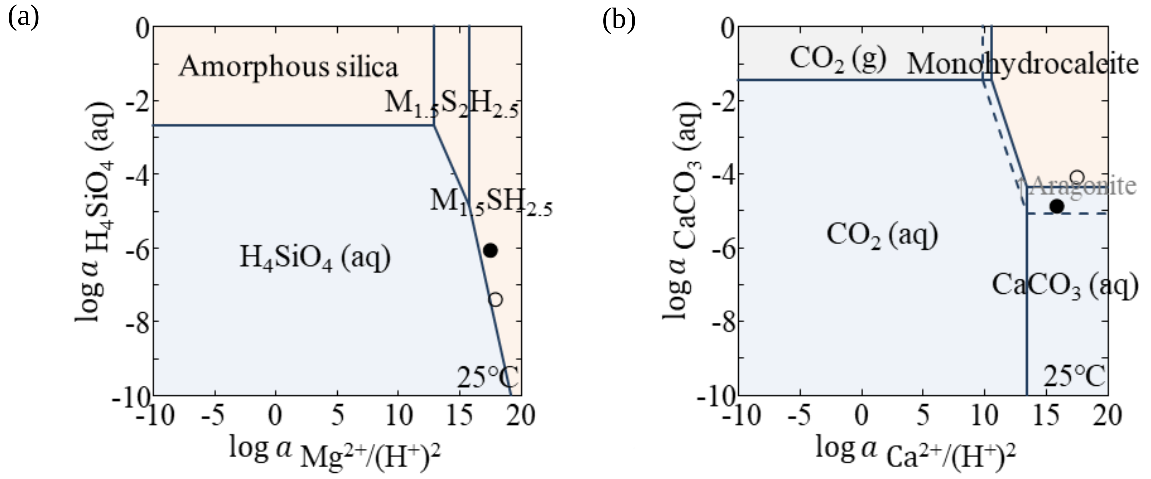

Figure 3. Phase diagrams and thermodynamics of effluent samples (solid and open symbols display effluent data from S1 and S2, respectively): (a) A phase diagram of Mg-silicates. $M_{1.5}S_2H_{2.5}$ and $M_{1.5}SH_{2.5}$ are M-S-Hs (formula: $(MgO)_{1.5}(SiO_2)_2(H_2O)_{2.5}$ and $(MgO)_{1.5}(SiO_2)(H_2O)_{2.5}$, respectively); (b) A phase diagram of Ca-carbonates. Dotted lines show stability boundaries of aragonite.

4. Discussion

From the thermodynamic calculation showed in section 3.2, the effluent is saturated M-S-H ($M_{1.5}SH_{2.5}$). This is consistent with results of the solid analyses (section 3.1). The both results confirmed precipitation of M-S-H (low-crystalline chrysotile) at ambient temperature (Figure 2 and Figure 3a). Moreover, aragonite was also precipitated probably via monohydrocalcite (Figure 3b), which is a metastable precursor. It is well known that dissolved Mg inhibits calcite formation, so that it promotes aragonite formation, although calcite is normally more stable phase than aragonite at ambient temperature. Therefore, under around pH 10, dissolved Ca and Mg consumed for aragonite and M-S-H formation respectively in Ca-Mg-CO_2-Si-H_2O system. In the system, dissolved Mg preferentially binds with dissolved silica, not with carbonate and hydroxide ions, although it is depending on activities of the ions.

In safety assessment of radioactive waste disposal, it is important to understand secondary minerals formed within the engineered barriers and at their interfaces. For this reason, many experimental studies for M-S-H formation have recently been conducted. However, the experimental products reported so far were so poorly crystalline that identification could not be done well (e.g, Nied et al., 2016). This was likely because saturation state of the dissolved Mg and Si were too high in comparison with natural reaction system, and the experiments were conducted in closed system. On the other hand, a few natural analogue studies have been performing field surveys under alkaline conditions. They suggest that secondary mineral (hydrated Mg-silicate mineral) is palygorskite (Milodowski et al., 2016) or trioctahedral smectite (Shimbashi et al., 2018), whereas this study confirmed chrysotile-like M-S-H, although duration time for formation and evolution is different between experimental and natural systems. Since geochemical/geophysical characteristics of the secondary mineral influence safety assessment of the engineered barrier, further investigations (esp. field surveys) need to clarify what factors specifically determine the formation of secondary mineral (Mg-silicate) species.

5. Conclusion

This study performed a field survey for the observation of present-day precipitation of M-S-H by mineral-water interaction at ambient temperature under alkaline conditions in the Kamuikotan tectonic belt, Hokkaido, Japan. TEM showed that the M-S-H was mineralogically identified as low-crystalline chrysotile, although XRD of bulk sample could not clearly discern peaks of M-S-H. Moreover, even in the condition including dissolved Ca and carbonate ions, the M-S-H was also precipitate although aragonite precipitation occurred simultaneously. This observation of M-S-H precipitation in nature will support to understand long term performance of the engineered barrier for radioactive waste disposal.

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Formation mechanism of Fe-oxide concretions on Earth and its analogous implication for alteration history in early Mars

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No abstract available.

Natural radiation effect on clay mineral properties: comparison with laboratory study

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4. Session III - Far-field studies I

Pre- and postglacial palaeoseismicity in Finland – Evidence of multiple bedrock fault reactivation events

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Key words; Paleoseismicity, earthquake, postglacial fault, reactivation, Finland

1. Investigation of Sub- and postglacial faults

Slow loading of horizontal stresses during the advance of the continental Scandinavian Ice Sheet (SIS) and rapid unloading of the vertical stresses during the melting stage forces the bedrock to readjust. The present-day uplift rates in Finland are up to 9 mm/a and the overall uplift has been nearly a km since the retreat of SIS ca. 13 ka ago.

In 2014 the Geological Survey of Finland and Posiva Oy jointly launched a country-wide LiDAR -based search for postglacial faults (PGFs), paleolandslides and other morphological features possibly related to postglacial seismicity. The project 'Postglacial faults and their dynamics (PGSdyn)' had three main goals: 1) to reassess the existence and spatial distribution of PGFs and other seismically-induced features in southern Finland, 2) to continue investigations in the vicinity of the known PGF zones in northern Finland, and 3) to improve understanding of the reactivation mechanisms, internal geometry and rupturing age(s) and the reoccurrence of late- and postglacial faults.

Analysis of LiDAR based digital elevation model was the backbone of the work (Palmu et al. 2015). Various digital and statistical tools were used to analyse the surface ruptures and spatial characteristics of the PGFs and landslides found around the faults. Selected faults were investigated using conventional geological and geophysical field methods (GPR, trenching, geological and Quaternary geological studies, drilling). The main conclusions of the PGSdyn –project are (Sutinen et al. 2014, 2019; Ojala et al. 2017, 2019a-c; Mattila et al. 2019):

(I) The PGF zones are characterized by a number of 0.2–9.1 km long fault scarp segments which form longer PGF systems. These systems further form PGF complexes which may be tens of kilometers long. By the end of year 2019, 143 segments, 18 systems and 9 complexes were identified from Finland.

(II) The PGF systems and complexes generally strike SW–NE directions and represent the reactivation of the pre-existing bedrock fault zones.

(III) Several of the excavated sites indicate that multiple rupturing events can take place within PGF complexes where different segments have been activated or reactivated at different times.

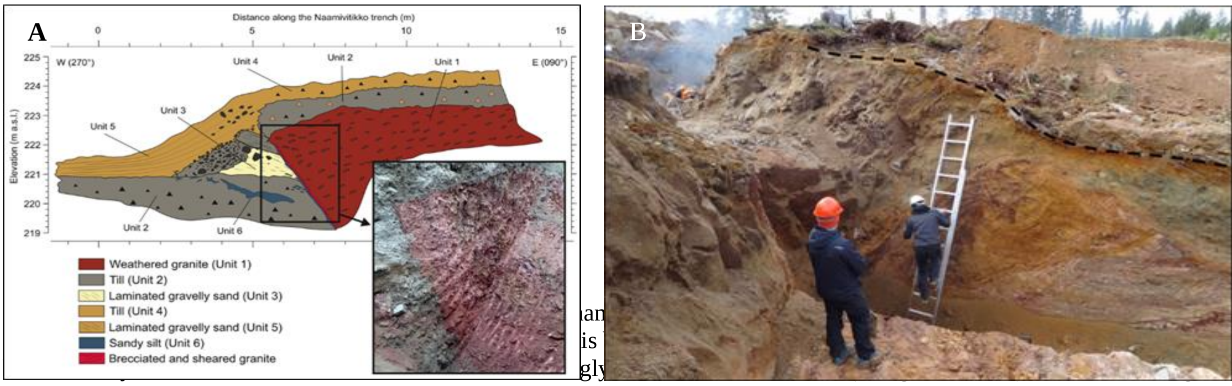
(IV) Field evidence show that seismicity has also continued throughout the Holocene and that the same PGF complexes were also active after glacial phases that predated the Late Weichselian glacial maximum. The landslide age data from northern Finland indicates that landslides associated with specific PGF system and complex may have different ages possibly suggesting cyclic seismicity.

(V) Realistic potential moment magnitudes for the earthquakes based on the rupture length, mean/maximum vertical displacement for PGF systems and landslide volume-area data are in the range of Mw ≈ 4.9–7.5 if assuming a single rupture event for each segment.

2. Relationship with ancient deformation zones

Altogether seven PGF segments in different geological setting and of different orientation were trenched in the Lapland area. One or more trenches were dug across fault scarps. It was attempted to expose both hanging wall and footwall and in most cases we succeeded to do so.

Without exception the PGF's we found to occur in strongly deformed and altered bedrock, within a single lithology unit or within more complex contact zone. Typically the alteration at the fault core was so intense that the excavator was able to dig through the fault plane without difficulty (Figure 1.). The quality of the rock improves with distance away from the scarp on both sides. Nevertheless, fracturing remains abundant and some signs of alteration could be observed tens of meters away. Depending on the age of reactivation relative to glaciation the fault scarp has been deformed by moving ice, meltwater flow and other surficial processes.



...an ... nch in ... is ... tured, ... gly ... s unit lower right is deformed and altered. The grey wedge on top of these units is till. The dashed line shows the gentle bend of the ground surface observable from LiDAR DEM.

Some of the postglacial faults in Finland have been drilled to study their nature and geometry in bedrock (Kuivamäki et al. 1998; Sutinen et al 2014, Ojala et al. 2017). The outcome has been similar at all sites. The hosting bedrock includes one or several deformation zones. Increased fracturing and abundant high temperature alteration (hematite, epidote) and mobilization of sulphides indicate that the bedrock has experienced shearing conditions prior to reactivation. Since postglacial reactivation occurs at low temperatures it results to brittle behaviour of the rock, i.e. re-opening of old fractures or grinding of the fracture surfaces, but formation of secondary minerals is unlikely (Sutinen et al. 2014). Therefore, the identification and dating of reactivation features is rather difficult. Common loss of core close to the traces of PGFs makes it challenging to assess the exact dip of the faults. Geophysical methods can be used to detect deformation zones, but it is not possible to differentiate the reactivated ones from the others. However, it seems to be likely that the reactivation rupture follows the deformation zone to depth. By combining the surface exposure of the PGF and intensely fractured core sections it was estimated that PGFs typically dip in the range of 45-65°.

It is almost a rule that PGF segments and systems cut geophysical anomalies and lithological contacts (e.g. Ojala et al. 2019). This indicates that the established relationship between postglacial reactivation and ancient deformation zones at our sites would be a general feature for PGF systems.

3. Evidence of multiple reactivation events

After detailed studies of several PGFs in trench excavations it is obvious that many faults have reactivated several times (Mattila et al. 2019; Ojala et al. 2017, 2019b, c). Because the dating of faulting or reactivation is particularly difficult in these systems, the multiple dating evidence is coming from sedimentological records of Quaternary sediments and landslides associated with the late- and postglacial fault zones. Textural features in till, the nature of an interface between different till units and bedrock and the general knowledge of the age of the till deposition allow to make interpretations of the dynamics of the fault system and thus, provide information on the relative timing of the fault rupturing events. However, since the sediment record is reset to great extent during each glaciation, it is challenging to 'date' the events beyond the Weichselian stage (~115 000–10 000 BP). It can only be speculated that the PGFs may have been active throughout the multiple Quaternary glaciations, or even before.

There were clear evidence that glacial till units of different ages at different stratigraphic position have or have not been affected by slip events and, on the other hand, bedrock fault scarps may have been deformed during deposition of basal till. For example, the Suaspalo trench in the Suasselkä PGF complex crosscuts two fault scarp segments (Ojala et al. 2019c) (Figures 2 and 3). The contact between lower till layer and the weathered bedrock is sheared due to the movement of ice during the deposition of the till unit. The pre-depositional fault scarps are smoothened and the soft material is dragged along the ice movement (Figure 2A). Interestingly, at the other scarp the shared contact is folded indicating younger, postglacial reactivation while the other segment has remained inactive (Figure 2B). Figure 3. shows the interpretation of the deformation history of the segments from Pre-Weichselian to postglacial time.

Repeated reactivation of postglacial faults is a geologically reasonable assumption, but to our knowledge it has not been thoroughly documented and confirmed before. The observations of the PGSdyn-project have important consequences for the discussion of moment magnitudes for the earthquakes related to postglacial faulting. As mentioned above, the potential moment magnitudes derived from rupture displacement data and landslide volume-area data are in the range of Mw ≈ 4.9–7.5. Considering the repeated reactivations it is likely that the surface expressions of some PGFs may be a result of more than one paleoseismic event suggesting that the individual earthquakes have been less severe than suggested by the rupture geometry.

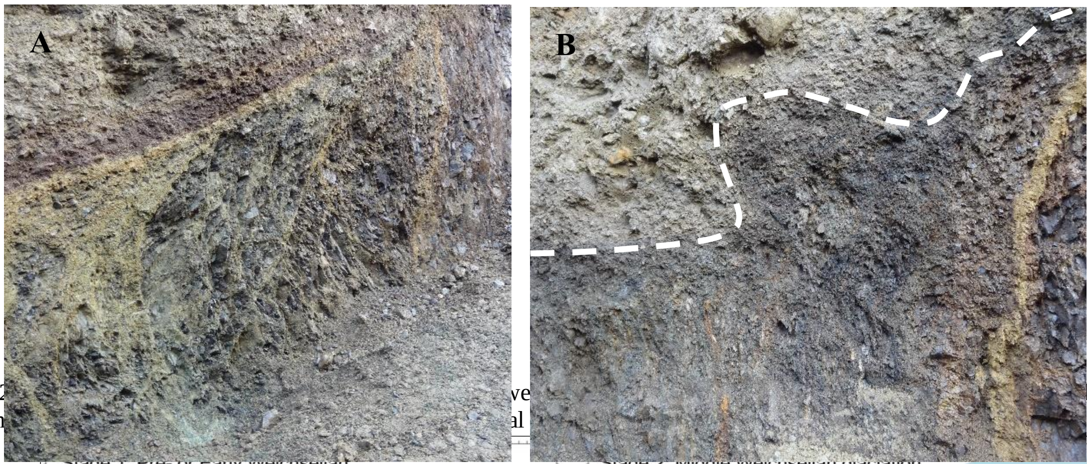


Figure 2 ... ve ... le at the other scarp th ... al

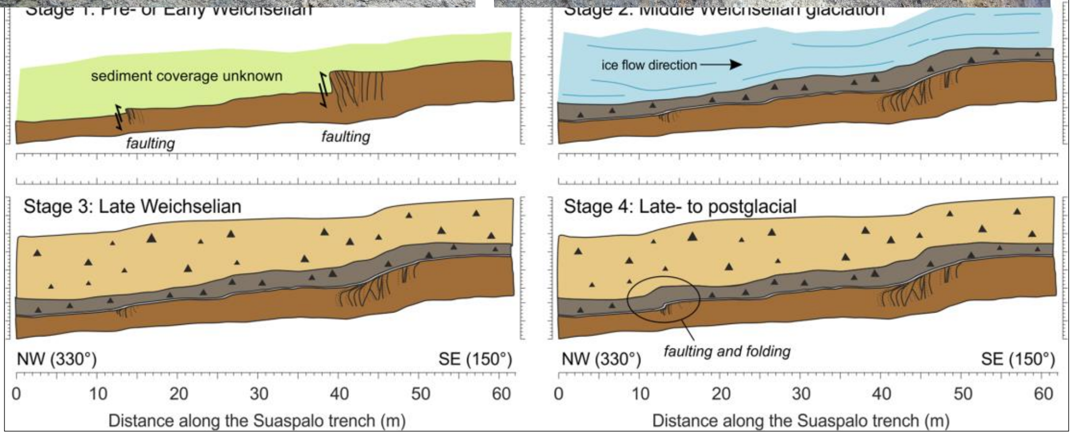


Figure 3. Development of faulting and reactivation features in the Suaspalo trench (Ojala et al. 2019b, c). Photo in Figure 2A is taken from the SE scarp and the photo in Figure 2B is from the NW scarp which has reactivated after till deposition.

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RWM Studies Relating to "Rock Matrix Diffusion" Using Natural Analogues

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No abstract available

The applicability of the Greenland Analogue Project (GAP) to the Finnish Safety Case

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Key words; ice sheet, permafrost, infiltration

1. Introduction

Within the safety case for geological radwaste disposal, safety assessments form a very important part to demonstrate compliance with regulatory constraints (dose or risk limits). In most safety assessments, uncertainties related to future developments are mostly handled by making pessimistic, and sometimes overconservative assumptions. This has been the case in the past, when assuming the consequences of glacial periods in the potential release and transport of radionuclides from the underground repositories to the surface environment. It is so, that previously it was e.g. assumed that infiltration of glacial meltwater could happen at all times when an ice sheet covered a site. The findings of the investigations of the GAP project have shown that this is not the case. Other lessons learned from the GAP project are applied to process understanding to add to multiple lines of evidence in arguing for the long-term safety and confidence building in nuclear waste management

2. Main findings of the GAP

The main findings of the GAP have been reported in Liljedahl et al. (2016). Here, those findings are first listed and then discussed on how they can be applied to the Finnish safety case.

The GAP study area, located to the east of Kangerlussuaq on the west coast of Greenland, covers approximately 12,000 km^2, of which about 70 % is occupied by the Greenland ice sheet. The ice sheet sits on the top of crystalline bedrock with similarities to the bedrock in the Fennoscandian Shield. The project focused into three main studies: 1) surface-based ice sheet investigations, 2) ice drilling and direct studies of basal condition, and 3) geological and hydrogeochemical studies beneath the margin of the ice sheet and permafrost (Figure 1).

Surface-based ice sheet investigations informed that surface melt and runoff happen in summer during at most 3 to 4 months. Ice drilling and direct studies of basal conditions informed on the basal thermal distribution, showing that most of the ice sheet from its margin to about 30 km inwards has a melted bed. Also, hydraulic measurements and analyses from the ice boreholes imply that ice overburden hydraulic pressure corresponds to 92% of the ice thickness. The combined results of these two studies show however that even if all surface melt and runoff reach the bed of the ice sheet, the meltwater is drained out from the ice sheet to form proglacial lakes that eventually outflow towards rivers and the sea (Mikkelsen et al. 2013) and does not contribute to potential infiltration. Geological and hydrogeochemical studies beneath the margin of the ice sheet and permafrost have shown a strong evidence of stable and conditions with limited groundwater flow at depths below 300 m. The length the permafrost extends beneath the ice sheet could not be established (Figure 1), but it certainly occurs beneath the ice sheet margin, which may contribute to the stability of groundwater flow at depth.

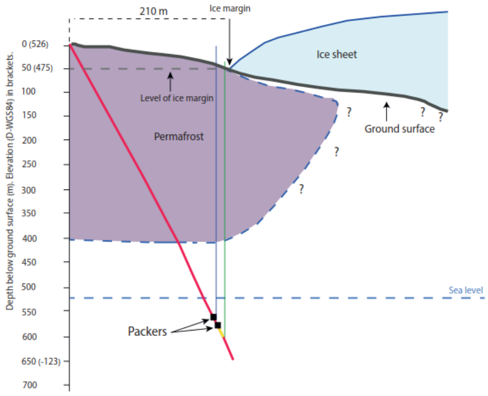

__Figure 1.__ Schematic illustration of the ice sheet (light blue), permafrost (purple) extent and deep borehole DH-GAP04. The vertical green and blue lines represent the head values at +485 m and at +520 m, respectively. The yellow colour shading along the borehole below the lower packer illustrates the approximately 30 m intact rock unit found at this depth in the borehole.

3. Applicability of the findings to the safety case

An immediate application is the setting of boundary conditions for hydrogeological studies during glacial periods, i.e. a hydraulic head corresponding to 92% of the ice thickness. Other applications are less immediate and more subtle, nonetheless contributing to the understanding of processes during permafrost and glacial conditions. They also contribute to understand the current conditions at the Olkiluoto site and to envisage how the evolving conditions after repository closure may affect the site and the repository. The fact that below 300 m depth the groundwater flow and chemistry is stable at both sites, Olkiluoto (Posiva 2013) and GAP site may not be a pure coincidence. Both sites have undergone several glaciation events, and signals of meltwater penetration is not found below 300 m depth. At the GAP site this is explained due to low storage capacity of the fracture system (Liljedahl et al. 2016), and at the Olkiluoto site this explained due to the buffering capacity of the bedrock and also due to the configuration of the water-conducting fractures, that would not allow the infiltration of meltwater to depths below 300 m.

The availability of meltwater during summer months is also a finding to be accounted for in groundwater flow modelling and groundwater chemistry. This was already taken into account in the previous safety case TURVA-2012 (Posiva 2012) when estimating the potential consequences of chemical erosion of the buffer bentonite due to intrusion of low-ionic strength meltwater.

The salinity of the borehole DH-GAP04 groundwater was relatively high regardless of their isotopic signatures show they originate from glacial meltwaters. In the drillcore it was observe that gypsum ($CaSO_4 \cdot 2H_2O$), the major fracture infilling mineral buffer contributes to salinity, as the Ca^{2+} and SO_4^{2-} concentrations show saturation with respect to this mineral. At Olkiluoto the major fracture infilling mineral is calcite ($CaCO_3$) that could contribute, if not to salinity, to the buffering of potential infiltration of meltwaters (e.g. Sahlstedt et al. 2009, 2014a,b). Also it could be argued that the occurrence permafrost inhibits infiltration, thus contributing to the resilience of the system to infiltration events, so that groundwater composition recovers from potential periodic infiltration of meltwater.

All in all, the lessons learnt from the GAP are used in the Finnish safety case to set appropriate and realistic boundary conditions for groundwater modelling and to argue on the conservativeness of the assumptions used in safety assessments.

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Fracturing in crystalline bedrock at different scales – An analogue for the Finnish repository site

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Keywords; Fracturing, Crystalline bedrock, Lineament, UAV-mapping

1. Introduction

One of many challenges of Deep Geological Disposal (DGR) of nuclear waste is the influence of fractures, faults and shear zones within the bedrock volume of the repository site and its immediate surroundings. The brittle structures at the repository site can affect the bedrock in several aspects e.g. seismic activity, groundwater flow and groundwater chemistry. To asses and minimize the risk from this aspects it is important to gain a profound understanding about the fracture/fault systems and the tectonic history of the site.

To increase this knowledge about brittle structures at all scales the 4-year project KYT-KARIKKO was initiated this year. The primary aims of the project is to enhance scientific understanding of the development and properties of fracture and fault systems in different geological environments and tectonic settings in southern Finland. The gathered data can be compared with data from the Finnish repository site in Olkiluoto and thus advance the geological modelling.

2. Plans and Outcome of the project

Our studies are targeted to areas with exceptionally well-exposed outcrops along the coastline of southern Finland. For these areas, new brittle data is collected on several different scales - regional lineament interpretation on the basis of geophysical and topographic data, fracture trace data based on UAV (Unmanned Aerial Vehicle) photogrammetric data and detailed-scale outcrop mapping. The analysis of the properties of fracture networks, such as fracture length, intensity and topology are mostly limited to 2D datasets.

For the topographic lineament interpretation, the data is interpreted in three different scales. For the UAV photogrammetric fracture trace mapping we use Phantom 4 Pro drone for taking photographs with flight altitude varying between 2-100 m depending on the purpose and required resolution. A flight altitude of 20 m has proved to be suitable for mapping relatively large areas and identifying most fractures over 1 m in length. To obtain data from the centimeter scale fractures we photographed smaller areas, in size of 5 x 5 m from a 3 - 5 m altitude. From the photographs we produce high-resolution orthomosaics and digital elevation models which are used for manual fracture trace mapping. Unfortunately any current automatic techniques for lineament and fracture identification have not yet provided us a better result than manual mapping. That could therefore be an interesting field for future research and thus a development into an automatic technique would remove interpretation variances and the main bottleneck for acquiring very large datasets.

The starting point of the comparison between the different scales is the evaluation of the fracture length and intensity distributions and the comparison of the distributions to the known scaling distributions of fracture systems (power law, exponential, gamma and log normal). If the fracture scalability relationships on different scales can be verified, then the fracture lengths and intensities can be predicted between different scales.

3. Preliminary results

The result from the first part of the study shows that brittle structures and scaling patterns can be investigated by combining multi-scale; LiDAR, airborne geophysics and UAV datasets (Figure 1). The scale between 100 m – 1 km is usually not possible to sample accurately but with these multi-scale studies it is possible to predict. UAV fracture mapping has emerged as an essential and efficient tool for characterise fracture networks, but it cannot substitute traditional structural geological mapping, merely complement it. The acquisition of this 2D data is vital to solve 3D problems and is an important input for DFN (Discrete Fracture Network) modelling. Essential parameters which are possible to acquire from the UAV fracture datasets are trace length, orientation, intensity, topology, kinematics and age relationships. The fracture trace maps also enable us to evaluate the influence of lithology, ductile precursors and major fault structures on the development of fracture networks.

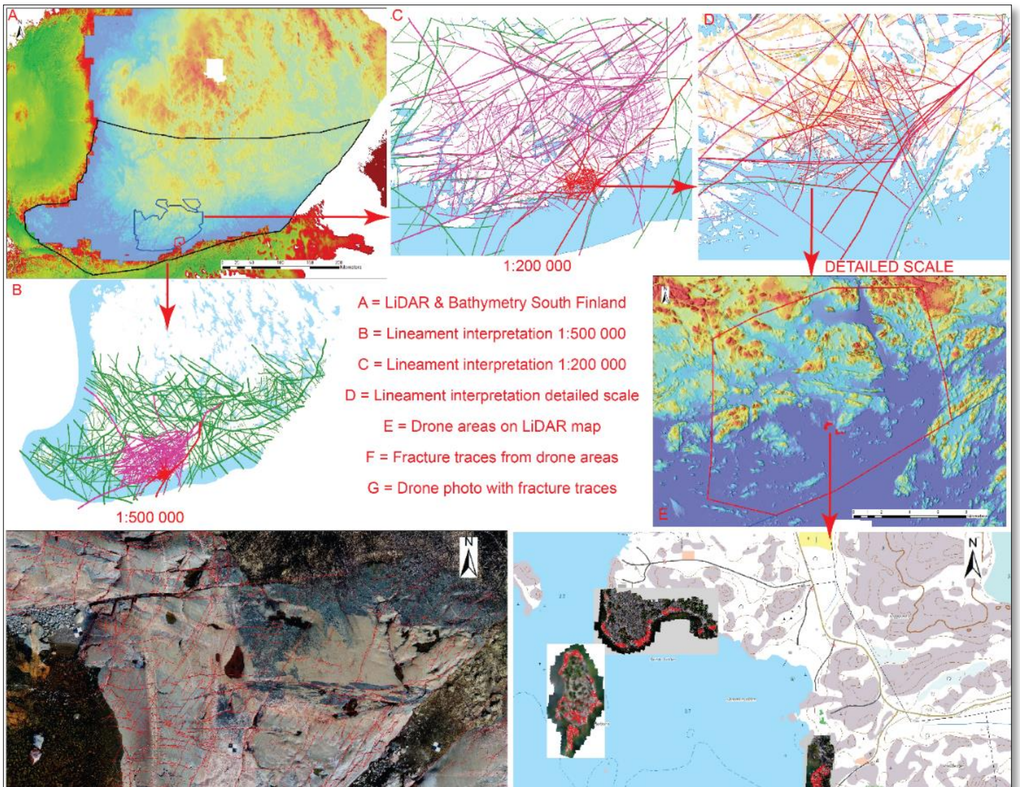

Figure 1. Multi-scale data acquisition, from lineament scale to detailed UAV scale.

4. Conclusion and Future plans

Within this project we will test different workflows for collecting and analysing datasets to define an efficient workflow for data collection and analysis. The main goal is to assess and mitigate the current uncertainties related to 3D and stochastic modelling of brittle structures. The results and research undertaken as part of the project aims at an advanced understanding of brittle tectonic events and the effect of brittle structures on e.g. rock mechanical processes and hydrological pathways in the bedrock.

A natural analogue study in KAERI: geochemical characteristics of deep geology in KURT

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No abstract available.

5. Session IV - Far-field studies II – self-analogues

Use of self-analogues in the Japanese SC

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No abstract available.

Long-term stability of fracture systems and their behaviour as flow paths in crystalline rock: an overview

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In granitic rocks, fracture networks typically provide pathways for groundwater flow and solute transport that need to be understood to assess the long-term performance of deep underground storage or disposal facilities such as radioactive waste repositories. However, relatively little is known about the long-term processes of fracturing and/or the longevity of flow paths (FP) in granitic rocks distributed within orogenic belts (e.g. Yoshida et al.2013). To clarify these issues, Japanese several plutons of different ages and in situ fractures in granite at the Mizunami Underground Research Laboratory (MIU) located in central Japan were studied. Detailed structural characterization and geochemical analysis of in situ fracture fillings sampled from up to a depth of 800 m were carried out to clarify the relationship between fracturing and mineral infilling processes.

The geometries of fractures and occurrences of fracture linings and fillings in different aged plutons reveal three stages of fracturing and mineral filling as follows. Different plutons show identical episodes of fracturing and fracture filling, consisting of: brittle tensile fracturing, due to decreasing temperature through the ductile–brittle transition after plutonic intrusion (Stage I); relatively rapid uplifting (ca. a few mm/year) accompanied by hydrothermal water circulation, which produced uncrushed layered mineral fillings (Stage II); and a period of low-temperature meteoric water circulation following exposure after uplift (Stage III) (Fig.1). In particular, structural features, mineralogical studies and dating of carbonate fillings suggest that stress has been accommodated by episodic widening of fractures formed early in the cooling history of each pluton, rather than by the creation of new major fractures in the intact part of the granitic rock. That is, the overall geometry of the fracture system has been stable throughout the history of the plutons following initial cooling through the ductile–brittle transition. Similarly, fractures that are orientated at a high angle relative to the minimum principal stress orientation have continued to host FP throughout the period of pluton uplift and associated fracture dilation. However, within such a fracture, the flow is channelized and the precise location of a FP within is believed to have changed over time in response to a combination of progressive calcite precipitation and episodic dilation. According to the model proposed here, mineralogical evidence for past fluid flow in granitic bodies within the circum-Pacific orogenic field can be used during geo-engineering to help interpret present patterns of groundwater flow. Additionally, future fluid flow pathways in these rock bodies are likely to have similar geometries as in the past. Therefore, future plate tectonic processes over timescales of millions of years may not fundamentally change the nature of groundwater flow pathways. These observations can be used when planning underground storage or waste disposal facilities within such rock bodies and when developing safety cases for such facilities.

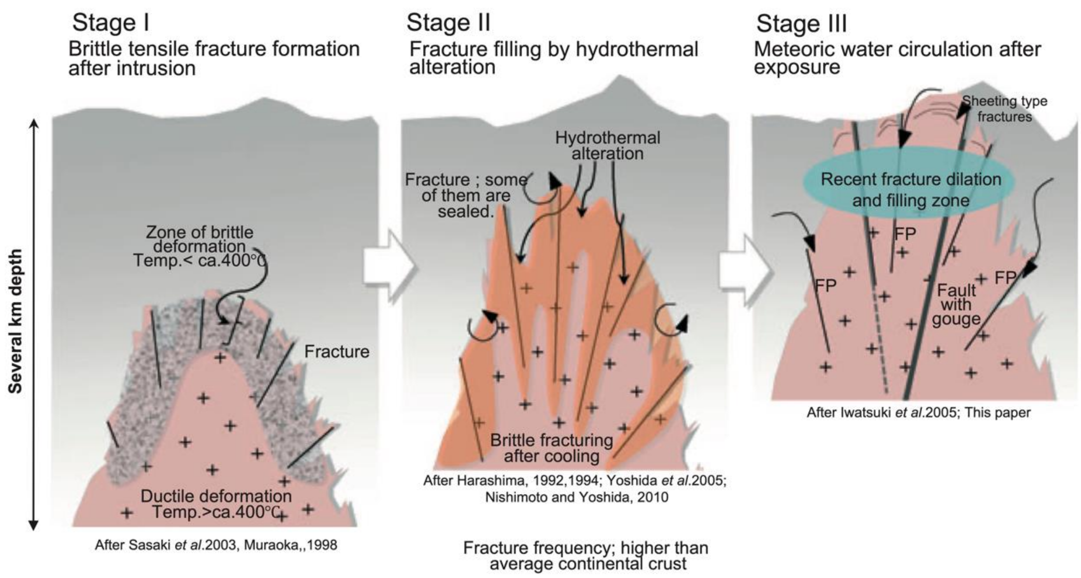

Fig.1 Schematic model of fracture formation during the evolution of a granite pluton within the Japanese orogenic field.

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Introduction to the KINA (Kiruna International Natural Analogue) project

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No abstract available.

6. Session V - The use of NA in stakeholder communications

Perception and communication of uncertainties by/with stakeholders: overview of a recent NEA meeting on Managing Uncertainty in Repository Siting and Implementation – Creating a Dialogue between Science and Society

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In 2017, the NEA Forum on Stakeholder Confidence (FSC) and the NEA Integration Group for the Safety Case (IGSC) held a Joint Workshop on Safety Case Communication. The NEA Working Group on Public Communication (WGPC) also participated. The workshop served as a platform to identify specific topics and working approaches for future collaboration between the working groups. One of the topics identified was "Managing uncertainty". This led to a second workshop (held on 9 October 2019), which specifically focused on this topic. A Programme Committee with members from the IGSC and FSC bureaux was responsible for the organisation of the joint workshop.

Toward Public Confidence on Long-term Safety of Geological Disposal

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Key words; Public Understanding, Confidence Building, Long-term Safety, Natural Analogue

"Nationwide Map of 'Scientific Features' relevant for Geological Disposal" was published by Ministry of Economy, Trade and Industry (METI) in July 2017. Nuclear Waste Management Organization of Japan (NUMO) together with METI has been holding nation-wide meetings since then and more than 100 meetings have been held until September 2019. In such meetings, communication in a small group has been introduced in order to enhance face-to-face communication and answer to all questions. During the meeting questionnaire surveys have been organized and the result shows that the communication in small groups is favorably received by the participants. However it shows that the confidence in safety of the geological disposal remains low. It shows that the way NUMO staffs communicate with the participants on safety needs further improvement.

In November 2018 NUMO published a "Safety case report" and peer review by Atomic Energy Society of Japan (AESJ) is on-going. One of the current challenges NUMO has been faced is how to explain the main outcomes and messages of the report to the general public. The NUMO safety case report was produced targeting experts on geological disposal and the contents are highly scientific / technical. It is therefore quite a challenge for non-technical public to understand the contents. Broad discussion is on-going to how NUMO could deliver the main outcomes and messages to the general public based on the NUMO safety case report.

The author has served as a communication officer in the NUMO's PR department for 5 years period (2014-2019). Through the period broad range of experience on communication with general public and young generation has been obtained. One of the critical findings through such experience was that the "Correct explanation and understanding on Safety" doesn't always guarantee "Confidence to safety of geological disposal". One of the reasons of such situation is that the written materials and explanations of safety by implementers are designed in a very "correct" manner to avoid any criticism from the opponent group. Therefore it tends to be rather "dry" contents and not always easy to understand materials (or explanations) for general public. Furthermore even if a person is somehow convinced on safe implementation of the geological disposal, he/she still doesn't feel comfortable accepting geological disposal nearby. It indicates that the understanding safety doesn't always link to building confidence.

During the presentation the author will introduce overview of the NUMO's communication (or PR) activities and share some of the critical experiences from public communication on safety. Then preliminary thoughts on how the communication on safety should be designed in order to obtain confidence to the geological disposal project will be explained. One of the promising candidates for such communication is an application of Natural Analogue (NA). There are, however, some critical questions to be answered if NA is used for public communication. The author expects to discuss using NA for communication on safety.

The use of natural analogues in building stakeholder confidence in Canada's plan for the long-term management of used nuclear fuel

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No abstract available.

Popularizing natural analogues: what a challenge! Some propositions from a PhD student in Epistemology

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Key words; Natural analogues; building confidence; long-term prediction; safety assessment of repositories.

1. Introduction

Natural analogues are a key element in assessing the performance and safety of repositories in the long-term (Chapman et al., 1984). But popularization is required to appreciate its contributions, because it is linked to complex concepts of different disciplines like radioactivity, the uniformity principle and reasoning by analogy.

Each stakeholder needs a clear idea of these concepts to have a chance to understand the value of natural analogues, even before talking of being convinced or not on the safety of geological disposal. How can one start a discussion if some arguments are incomprehensible? Popularizing gives the public the opportunity to make a more informed decision.

My aim is to introduce natural analogues as a field of research in my discipline, because it is not referenced. It means demonstrating that it could be an object of research, finding an approach to study it and indicating what it brings to Epistemology, especially through reasoning by analogy in science. For the NAWG-16 workshop I will put the emphasis on my recent research: The contribution of epistemology to the popularization of natural analogues.

2. Methods

The explanation of natural analogues should be deep enough to make understanding possible and build confidence. It is a huge challenge and maybe only non-specialists can confirm the success of popularization. I have an ideal background, with notions of physics, chemistry and geology from *Lycée* (High School) and a few elements on radioactivity and popularization. So, I began with myself!

I started to read scientific papers in English, especially Ewing's works (Ewing and Haaker, 1979) and proceedings of the Oklo Working Group and the Natural Analogue Working Group. To have a thorough understanding and a common vocabulary, scientists active in this field, or retired, were interviewed and continue to be.

I have had a few opportunities to test different ways of explaining natural analogues through posters and during public conferences, for example in the *Palais de la Découverte,* a French Museum and Scientific Cultural Center, where that was my mission. I look forward to testing the following propositions.

3. Propositions

After five years of research and attempts to understand this little known concept in my field, starting with the following questions is crucial:

i. Why is it necessary to predict over the long term?
ii. How to attempt a long-term prediction?
iii. What are the “scientific” results through natural analogues?
iv. And how does it influence the design and building of a repository?

These questions include complex concepts mentioned before (radioactivity, uniformity principles and reasoning by analogy) that need to be explained.

In addition, the defence of reasoning by analogy (Petit, 1992; Ewing, 1995) could be more consistent by referring to the “Plea for the High Status of Natural History” by Stephen Jay Gould (Gould, 1990) and by specifying the question of relevance; which depends on the subject, the historical context and logical details (Bartha, 2019). The value of reasoning by analogy has evolved over time (it also depends on the discipline to which we refer), so this work is crucial.

I also argue that each natural analogue has to be studied, to distinguish the natural analogues used as metaphors and the methodology of long-term prediction using a specific natural analogue. This is because analogy is ambiguous, as it accepts several interpretations. The general criteria proposed have to be refined, especially “the chemical analogy, [which] should be good” (Chapman et. al, 1984), to check the robustness of the reasoning by analogy and to build confidence.

This challenge also involves, as does every attempt at popularization, detecting and stating elements that are obvious to the specialist. For example, I ask the question; “why is there a focus on water?” Water infiltrates the rock and destroys materials used to confine high-level radioactive waste. Another point is to question the traditional categories of stakeholders, such as experts, politicians and the public, in the light of developments in information technology. Who doesn’t use the internet? Should not categories based on individual knowledge relevant to popularization?

Acknowledgements

I would like to thank Russell Alexander for the opportunity to present my research to specialists in this field. I would like also to thank Heini Reijonen and, again, Russell Alexander for listening to my efforts to create a bridge between epistemology and natural analogues.

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7. Session VI - Microbiology in the bentonite buffer

Microbial populations in natural bentonites: results from IBL

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No abstract available.

Biotransformation of trivalent actinides - effects of halophilic and denitrification microorganisms-

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Key words; TRU waste, HLW, co-location disposal, TRLFS, coordination environment diagram

1. Introduction

Community analyses of microorganisms indicate that the microorganisms inhabit in the different adequate environments (Selenska-Pobell, 2002). The microorganisms which adapted to the local geochemical conditions were well grown. These microorganisms function a major role in the mobility of radionuclides in the environment through many processes including sorption at the cell surface, precipitation (Martinez, 2007) and redox reactions (reduction and oxidation) (Dispirito and Tuovinen, 1982, Lovley et al., 1991). If microorganisms are planktonic, they may enhance the migration of radionuclides by sorption. If microorganisms form a biofilm, they may immobilize radionuclides.

In Japan, TRU waste containing large amount of nitrate salt plans to dispose with high level waste (Co-location disposal) (Mihara et al., 2011) in geological formation. When we estimate the impact of the co-location disposal, groundwater flow paths like those occurring in mineral filled fractures, wherein nitrate may disperse from a repository to groundwater. Thus, groundwater is characterized by high nitrate concentration and high salinity. Sorption of radionuclides involving TRU and FPs by halophilic microorganisms and denitrifying microorganisms should be studied.

We have conducted the sorption of trivalent actinides (alternatively used Eu(III)) on the cell surface of halophilic microorganisms, *Halomonas* sp., *Halobacterium salinarum*, *Halobacterium halobium*, (Ozaki, et al., 2004, Ozaki et al., 2005) and on the cells of denitrifying microorganisms, *Paracoccus denitrificans* (Ozaki, et al., 2006). Chemical species of the adsorbed Eu(III) were analyzed by time-resolved laser-induced fluorescence spectroscopy (TRLFS).

2. Sorption of Eu(III) by halophilic microorganisms

Microbial cell surface is constructed of a range of organic acid functional groups that can adsorb metal cations and protons from solution. Functional groups on microbial cells surface are similar to those found on mineral surfaces in that they are proton active. Subsequently most microbial cell surface functional groups are positively charged in low pH, and neutrally or negatively charged by increasing pH. Even though the functional groups are dominant adsorption sites of cations in microbial cell and minerals, the properties of the functional groups are different. In microbial cell functional groups consist of carboxylic, phosphoric, amine sites, and proteins. In the silicate minerals, those are silanol and aluminol functional groups.

Chemical species of the adsorbed Eu(III) were analyzed by time-resolved laser-induced fluorescence spectroscopy (TRLFS). Figure 1 shows that the relationships between $R_{E/M}$ and ΔN_{H2O}, which is called hereinafter as coordination environment diagram, obtained for the microbial species. In Figure 1 ΔN_{H2O} shows number of water molecules in the inner-sphere of Eu(III), calculated according to the equation $\Delta N_{H2O} = 9 - N_{H2O}$. The relative peak intensity ratio ($R_{E/M}$) is defined by the ratio expressed by $R_{E/M} = I(^5D_0 \rightarrow {}^7F_2) / I(^5D_0 \rightarrow {}^7F_1)$, where $I(^5D_0 \rightarrow {}^7F_2)$ and $I(^5D_0 \rightarrow {}^7F_1)$ are calculated from the peak areas at 614 nm ($^5D_0 \rightarrow {}^7F_2$) and 592 nm ($^5D_0 \rightarrow {}^7F_1$), respectively.

Coordination environment diagram shows that an almost comparable ΔN_{H2O} was observed for *Halomonas* sp. and *P. fluorescens*, while the $R_{E/M}$ for *Halomonas* sp. was larger than that for *P. fluorescens*. This finding suggests that the structure around the functional groups for Eu(III) on *Halomonas* sp. was more complicated than that on *P. fluorescens*.

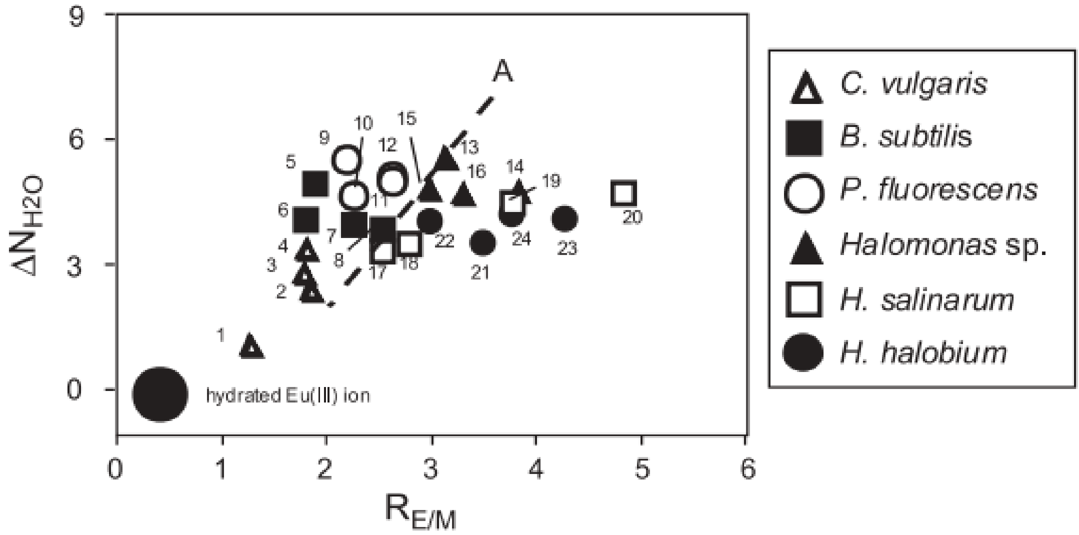

Figure 1 Coordination environment diagram obtained for the microbial species. The large closed circle represents the result obtained for hydrated Eu(III) ion with no interaction other than water molecules. pH values are as follows: (1) 2.8, (2) 3.9, (3) 4.4, and (4) 5.8 for *Chlorella vulgaris*, (5) 3.3, (6) 3.8, (7) 5.0, and (8) 5.5 for *Bacillus subtilis*, (9) 3.2, (10) 4.0, (11) 5.3, and (12) 6.1 for *Pseudomonas fluorescens*, (13) 3.7, (14) 4.5, (15) 5.3, and (16) 5.5 for *Halomonas* sp., (17) 3.6, (18) 3.8, (19) 5.2, and (20) 5.8 for *Halobacterium salinarum*, and (21) 3.5, (22) 3.7, (23) 4.9, and (24) 5.7 for *Halobacterium halobium*. (Ozaki et al., 2004).

3. Sorption of Eu(III) by denitrification microorganisms

Paracoccus denitrificans which grows both aerobically and anaerobically, and it anaerobically reduces a fixed nitrogen oxide. For comparison of sorption of Eu(III), we also studied the association of Eu(III) with Gram-negative bacteria, *Alcaligenes faecalis*, and *Shewanella putrefaciens*.

The kinetics study showed that the Eu(III) adsorption on the bacteria rapidly proceeded. The Eu(III) adsorption on *A. faecalis* and *P. denitrificans* at pHs 3, 4, and 5, and that on *S. putrefaciens* at pHs 4 and 5 reached a maximum within 5 minutes after contact. For *P. denitrificans*, the percent adsorption of Eu(III) decreased after the maximum percent adsorption was attained, which suggests the existence of exudates with an affinity with Eu(III).

Coordination environment diagram obtained for *P. denitrificans*, *A. faecalis*, and *S. putrifaciens* indicate that $R_{E/M}$ for *P. denitrificans* is larger than that for non-halophilic microorganisms and comparable to that observed for Eu(III) adsorbed on halophilic microorganisms shown in Figure 1. This suggests that the structure of the coordination site for Eu(III) on *P. denitrificans* is similar to that of halophilic Gram-negative bacteria, such as *Halomonas* sp., implying its adaptability to saline conditions.

4. Role of halophilic and denitrification microorganisms on the migration of trivalent actinides

Presence of microorganisms in/around co-location disposal site affects chemical species transformation of trivalent actinides. Around co-location disposal site, groundwater is characterized high nitrate concentration and high salinity. Such high nitrate concentration may enhance activity of halophilic and denitrification microorganisms. The kinetics study of the biosorption showed that the Eu(III) adsorption on the bacteria rapidly proceeded. Coordination environments of biosorbed Eu(III) by *Halomonas* sp. and *P. denitrificans* showed more complicate association with non-halophilic and non-denitrification microorganisms. These results suggest that migration behavior of trivalent actinides around the co-location disposal site of TRU and HLW is different from that around the disposal site for HLW alone.

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Laboratory and in-situ examinations of microbial abundance, composition and activity in compacted bentonite buffer materials

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Key words; microbiologically influenced corrosion, Kunigel V1, Horonobe Underground Research Laboratory

1. Introduction

Spent nuclear fuel (SF) and other high-level radioactive wastes (HLW) from nuclear energy production must be stored safely for at least 100,000 years. The safety requirements for HLW and SF disposal call for metallic containers to remain intact for a long period of time. Microbial activity in bentonite buffer materials in deep geological repository is of concern for microbiologically influenced corrosion which could affect the longevity of metallic containers. In a future repository environment, dominant corrosive elements for metallic containers will be oxygen and sulfide. Oxygen will be introduced during repository construction and microorganisms will contribute to consume it soon after repository closure. However, after the oxygen is reduced, it has been considered that sulfate reducing bacteria (SRB) would be able to survive and produce sulfide by performing the respiratory reduction of sulfate under anaerobic condition. Recent studies have revealed that not only SRB but also methanogens, acetogens, etc. can contribute to microbiologically influenced corrosion (MIC) (Dinh et al., 2004; 2018; Mand et al., 2014) (Figure 1). Previous reports of laboratory and in-situ experiments considering microbial activity in compacted bentonite have found that microbial activity is correlated with bentonite buffer density and its resulting water activity and swelling pressure (e.g. Stroes-Gascoyne et al., 2010; Masurat et al., 2010). In Japan, data has been collected regarding microbial corrosion of carbon steel or cupper with compacted Kunigel V1 bentonite. From the results of the experiments, it was evaluated that MIC would not affect the longevity of the containers, as SRB were not detected in the buffer materials (JNC, 2000). However, population of naturally occurring microorganisms in subsurface environments is different from those in the experiments, as isolated sulfate reducing bacteria, iron-oxidizing bacteria, and indigenous microorganisms in the bentonite were used for the corrosion experiments. It might be possible that naturally occurring microorganisms would have a better chance to survive and be active in buffer materials. The incubation period was only for a few weeks, maximum for 3 months, although it is known that growth rate of microorganisms, especially anaerobes, in natural environments is generally slow and they would take a few months or more to grow. In addition, culture dependent methods were used to detect microorganisms, although most of microorganisms (>99%) in natural environments are not be able to culture. There is not enough data using Kunigel V1 collected under Japanese geological disposal concept. To evaluate microbial effects on corrosion of metallic containers and activity in the bentonite buffer materials with Kunigel V1 and silicate sand, we have conducted laboratory-scale and in-situ experiments.

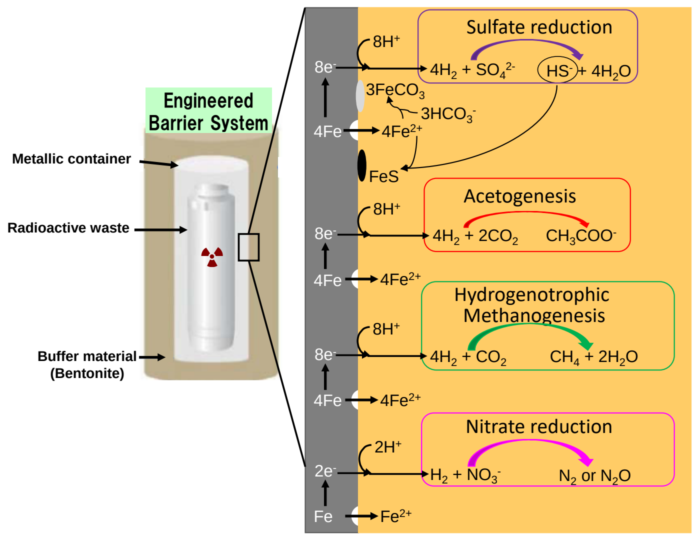

Figure 1 Conceptual scheme of microbial effects on corrosion of metallic container in geological disposal systems

2. Laboratory experiments

Corrosion of carbon steel buried in buffer material was investigated in the lab experiments using anaerobic corrosive microbial consortium described previously (Hirano et al., 2015). The buffer material was composed of 70% Kunigel V1 bentonite and 30% silicate sand with different dry densities of 1.0, 1.3, and 1.6 Mg/m^3 in test cells, and incubated with or without inoculation by an enriched microbial community for one year at 30°C and 50°C. Microbial viability was analyzed by culture-dependent technique, and microbial growth and changes in microbial population were analysed by culture-independent molecular techniques during the experiments.

Based on the weight loss measurements, corrosion rates were suppressed (9.4 – 12.9 $mg/cm^2/year$) in samples with compacted densities of 1.3 and 1.6 Mg/m^3 at 50°C. In contrast, significant corrosion acceleration by microorganisms was observed between 6 months and 1 year with the density of 1.0 g/cm^3 at 30°C. The corrosion rate was 52.0 $mg/cm^2/year$ after one year, although the corrosion rate was 3.2 $mg/cm^2/year$ until the sixth month. It is demonstrated that a sufficiently high dry density is one of the important key factors to suppress microbial activity in buffer materials surrounding metal containers, because of the physical characteristics such as small pores, low water activity, less nutrient supply caused by low hydraulic conductivity.

The results of microbial growth analyses using culture-independent molecular techniques showed that sulfate reducing bacteria and methanogen proliferated after one year at all densities of compacted buffer materials compared to the initial microbial densities. As a factor for the microbial growth in the compacted bentonite material, microorganisms might have been able to survive in inter-aggregate pores (macro-pores), as it has been reported that the micro-pores are formed inside MX-80 bentonite/sand mixtures with the size in the range of from 10 μm to 50 μm depending on the dry density (Wang et al., 2013). In case of 100% of MX-80 bentonite, the size of pore after swelling was distributed in the range of 0.005 to 0.1 μm in the mean size of 0.02 μm (Stroes-Gascoyne et al., 2010; Wang et al., 2013). In general, the minimum size of microorganisms is considered to be 0.2 - 0.3 μm, which is reasonable in terms of the size of biomolecules (cell membranes, proteins, ribosomes, genomic DNA, etc.) that are necessary to sustain life. However, some microorganisms, even if they have a size diameter of 0.2 μm or larger, can pass through 0.1 or 0.2 μm pore size doe to the flexibility of their cells (Wang et al., 2008), or change their size depending on environmental conditions (Kuhn et al, 2014). However, those microbial sizes and ecologies are special cases, and even if we take them into account, microbial growth is expected to be suppressed when the pore size of the buffer material is smaller than 0.1 μm. A previous report showed that pore size after swelling with a dry density of 1.6 Mg/m^3 using 100% MX-80 bentonite was distributed in the range of 0.005 - 0.1 μm (Stroes-Gascoyne et al., 2010). Results suggested that a low water activity (<0.96) and a swelling pressure >2 MPa appear to suppress microbial activity in bentonite buffer materials, which needs to be maintained at dry densities of 1.6 Mg/m^3 or higher in case of 100% MX-80 bentonite (assumed Effective Montmorillonite Dry density; EMDD of ≧1.4 Mg/m^3). The higher swelling pressure, the lower water activity in buffer material and smaller pore size, which is expected to be an extremely harsh environment for microorganisms to live in. The EMDD calculated with reference to Sato et al., (2009) is approximately 1.0 Mg/m^3 at a dry density of 1.6 Mg/m^3 with a mixture of 70% Kunigel V1 and 30% silica sand, in case of the montmorillonite content in Kunigel V1 is 0.59. Therefore, the swelling pressure and water activity conditions may not be sufficient to suppress microbial activity when a mixture of Kunigel V1 and silica sand is used as a buffer material. Although our results also confirm that temperature and high dry densities are important factors to suppress microbial activities, it is necessary to determine the condition to suppress corrosion rate of carbon steel and microbial activity with Kunigel V1, including in-situ experiments under subsurface environments.

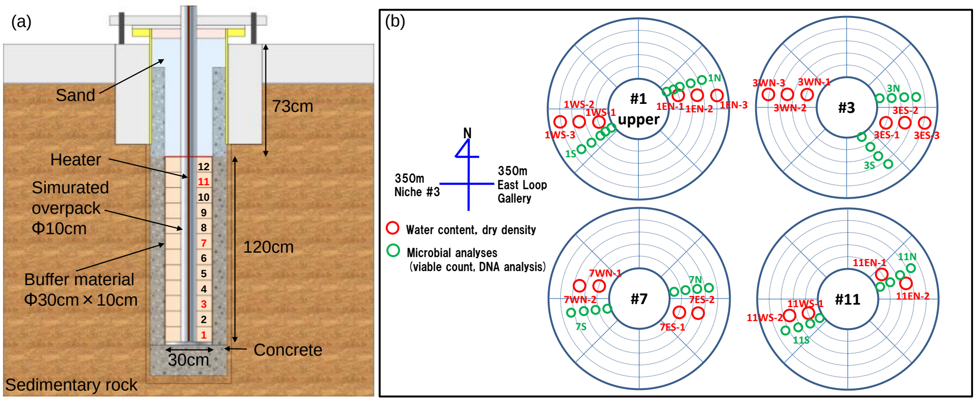

Figure 2 **Schematic drawing of the in-situ experiment for overpack corrosion at a depth of 350 m in Horonobe Underground Research laboratory. (a) Configuration of the OP experiment. (b) Sampling location in the bentonite buffer material.**

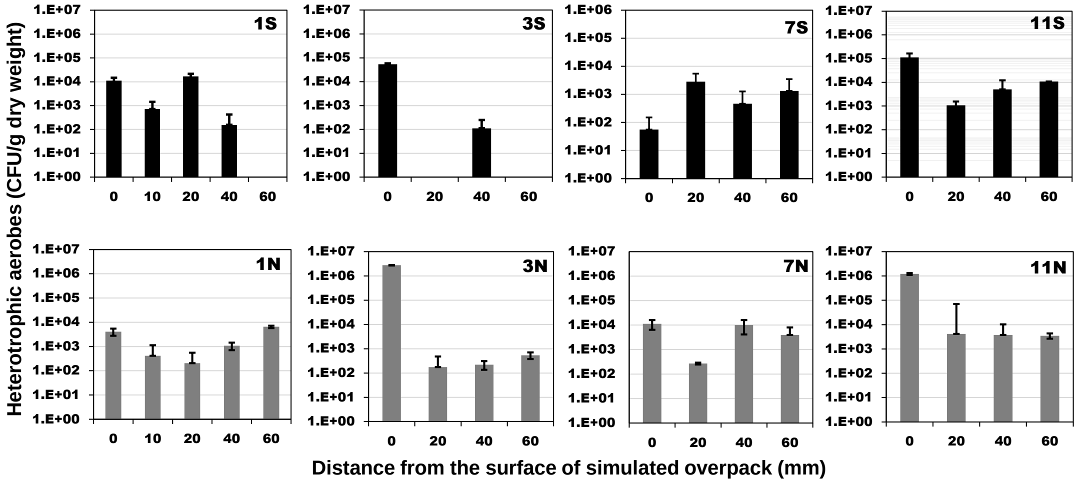

Figure 3 Culturable aerobic heterotrophic microorganisms in the buffer material

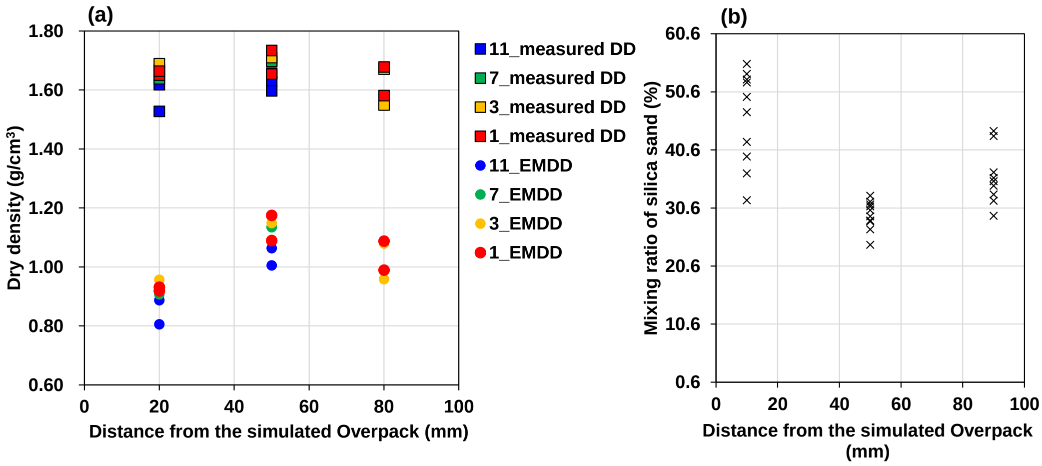

Figure 4 Physical properties. (a) Measured dry densities (DD) and calculated Effective Montmorillonite Dry Densities (EMDD) in the buffer material. (b) Mixing ratio of silica sand in the buffer material.

3. In-situ experiment at Horonobe URL

An engineering scale of in-situ corrosion experiment was set up and operated in sedimentary rocks at 350 m gallery of the Horonobe Underground Research Laboratory (URL), Hokkaido, Japan (Figure 2(a); Nakayama, 2018), in which various aspects of compacted bentonite buffer material and corrosion of carbon steel were studied. Buffer material was used a mixture of 70% Kunigel V1 bentonite and 30% silica sand, pressed into blocks (dry density 1.6 Mg/m^3). A heater inside the simulated overpack (OP) was maintained at 95 ºC for approximately 3.5 years until dismantling occurred. To assess microbial existence and population in the compacted bentonite buffer material, samples were collected after 5-year incubation at the subsurface environment (Figure 2(b)). Results showed that culturable aerobic heterotrophs in the buffer were heterogeneously detected in almost all the samples, at most 1.2 x 10^6 CFU/g (CFU: colony forming unit) dry weight at an interface location between the simulated OP and the buffer material (Figure 3). Previous reports of in-situ experiments using Kunigel V1 and an ore of Kunigel V1 showed that the numbers of aerobes in buffer materials were much lower compared to the number in this study, with the range of 6 x 10^1 – 1 x 10^3 (Stroes-Gascoyne, 2010; Aoki et al., 2010). It is assumed that the high densities of viable aerobes in this study was caused by large amount of silica sand intruded into the gap between the OP and the buffer material from the top of the buffer material unexpectedly. As a result, it is inferred that the silica sand became a water channel and many microorganisms got into the gap with groundwater. This phenomenon can be confirmed by the fact that goethite was one of the main corrosion product on the OP surface. Magnetite and siderite were also detected as major corrosion products on the OP surface. The presence of these minerals suggests that the circumstance in the buffer material had been changed to anaerobic condition as these minerals are mainly generated under low oxygen pressure. It is assumed that aerobic microorganisms could contribute to consume oxygen introduced with silica sand. The values of measured dry densities were detected in the range of 1.55 to

1.73 Mg/m^3 (Figure 4(a)). The EMDD were calculated using the measured dry densities and measured ratio of silica sand (Figure 4(b)) in the buffer material with referred to Sato (2009), and the values were in the range of 0.81 to 1.17 Mg/m^3. The results showed that measured dry densities and the EMDD varied heterogeneously (Figure 4(a)). The distribution of viable microorganisms in the buffer material seems to be caused by nonuniformity in dry density, and water content and swelling pressure etc. associated with the dry density in the transition period of the swelling process for the engineering scale experiment. These results may also demonstrate that it is very important to minimize heterogeneity of dry densities within buffer materials in repository environment. On the other hand, no anaerobic microorganisms were detected in any of the samples during the one-month incubation period by the culture method. Although the values of aerobic heterotrophs detected by the culture methods indicate the number of surviving microorganisms in the samples, these microorganisms may not indicate the number of microorganisms actively living in the environmental conditions, because it is known that some of the microorganisms were dormant, such as those that formed spores. It is possible that microorganisms did not proliferate in the buffer material and remained dormant after the consumption of oxygen and swelling of the buffer material, as the temperature in the proximity of the OP was higher than 90 ºC due to the heater installed in the OP. Further analyses by culture independent methods, such as DNA analyses, should be necessary to understand microbial distribution and roles effected on corrosion of the carbon streel material.

This study was performed as a part of "Project on Research and Development of Spent Fuel Direct Disposal as an Alternative Disposal Option" and "The Project for Validating Near-field System Assessment Methodology in Geological Disposal System" funded by the Ministry of Economy, Trade and Industry of Japan.

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8. Session VII - Introduction to the International Bentonite Longevity (IBL) project

International Bentonite Longevity (IBL) project: an introduction (description of the Tsukinuno bentonite mine)

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This extended abstract describes the preliminary results of a natural analogue (NA) study of the long-term behaviour of various aspects of the industrial bentonites planned for use in repositories for radioactive wastes. This work has been conducted within the International Bentonite Longevity (IBL) project which includes RWM Ltd (UK), NWMO (Canada), NUMO (Japan), KAERI (ROK) and GTK (Finland) as Project Partners. The work has been carried out in Kunimine Industries Company (KIC)'s Tsukinuno (NW Japan) sub-surface bentonite mine, a source of Miocene age Na-bentonite (at depth in the mine) and Ca-bentonite (near-surface). The site (Figures 1 and 2) is ideal for studying processes of direct relevance to safety cases (SC) for radioactive waste repositories that utilise bentonite as part of their multi-barrier safety systems (i.e. buffer, backfill and tunnel and borehole plugs and seals).

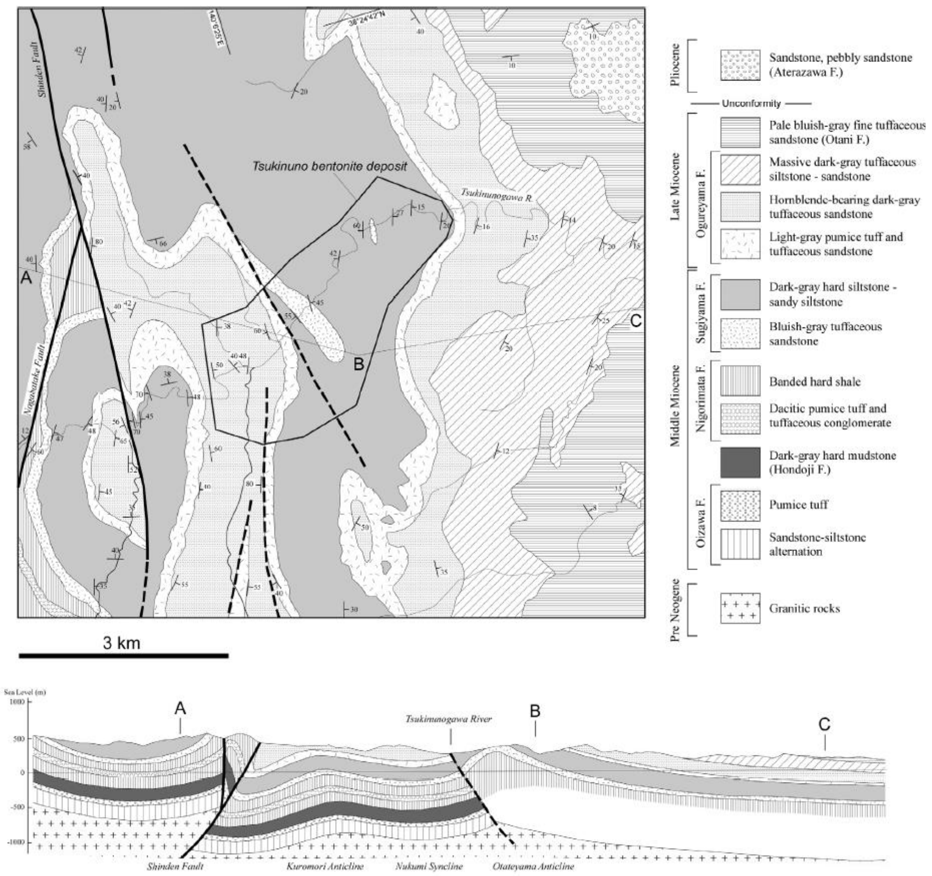

Figure 1: *Geological map and cross-section of the Ohe region, Yamagata Prefecture, Japan (Yamaji et al., 1986). KIC's Tsukinuno mine is located in this area.*

The IBL project focusses on studying long-term, safety-relevant processes:

- bentonite longevity;
- bentonite deformation (fracturing, swelling and heave);
- bentonite erosion (including bentonite-water interaction processes with fresh and deeper groundwater chemistries);
- suppression of microbial activities in bentonite.

Other areas of relevance were identified as:
- bentonite saturation state;
- bentonite sampling/analytical development;
- stakeholder communications.

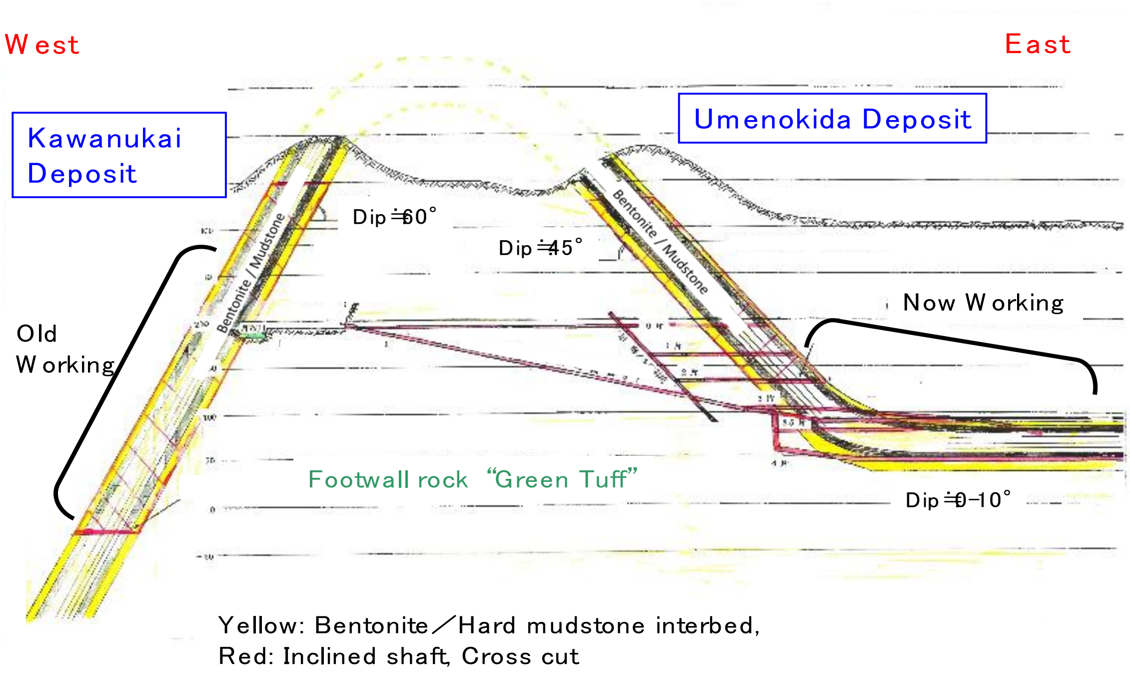

Figure 2: *Cross-section of the Tsukinuno mine, Yamagata Prefecture, Japan (image courtesy KIC).*

This document briefly introduces the project and presents the main conclusions of the IBL project Phase A (Reijonen & Alexander, 2021). These are:

Bentonite longevity. Although clear changes in the Cation Exchange Capacity (CEC)/Exchangeable Cation Composition (ECC) of two bentonite beds were observed, no other major changes (e.g. in bentonite mineralogy, geochemistry or structure) in the bentonites were observed. Further proof of the bentonite longevity should be obtained in IBL Phase B by examining the bentonite physico-chemical properties and by dating the period of ongoing groundwater-bentonite interaction. A preliminary study of natural pelletal bentonite indicated the value of a future, more detailed study of this material to provide supporting information on the long-term behaviour of industrial pellet bentonite which may be used in borehole seals.

Bentonite deformation. A preliminary examination on a zone of bentonite faulting indicates the presence of very old, layer-parallel shear. Of more relevance to bentonite seals is the identification of an associated fault which shows clear brittle fracture in the shale host rock is associated with ductile deformation in the neighbouring bentonite. This will be investigated further in Phase B of IBL as this is of direct relevance to the study of borehole seal-relevant fracturing in and around a seal.

Bentonite erosion. At least four environments were identified where bentonite erosion studies could be conducted. However, successful (i.e. artefact-free) sampling is extremely difficult, so time should be spent in considering appropriate approaches to the assessment of bentonite erosion in situ before any attempt is made to embark on such work in the identified environments.

Suppression of microbial activities in bentonite. Preliminary data are available (and are presented in Kremer, 2021 and Beaver et al., 2021) and these show very low microbial populations. These results are a NWMO in-kind contribution to the IBL project and will be fully available in Phase B.

Other areas. Due to limitations of time and effort (and the impact of COVID-19 in 2020/2021), other areas of relevance were not evaluated during Phase A of IBL. However, an initial assessment of those areas of direct relevance to the borehole sealing project indicates that appropriate sites exist in the Tsukinuno mine to study the main areas (i.e. bentonite saturation state, bentonite sampling/analytical development and stakeholder communications) of interest and this will be pursued further in Phase B of IBL.

The output from IBL Phase A has provided, in part, evidence of the longevity of bentonite under conditions of relevance to borehole seals. Further, it has acted as a proof-of-concept for the direct evaluation of borehole sealing-relevant processes in natural bentonites as NAs of the industrial bentonites likely to be used in future repository programmes.

Acknowledgements

Phase A of the IBL project was funded by RWM Ltd (UK) as part of their ongoing Borehole Sealing project and the authors would like to thank RWM for their support along with our colleagues in the Borehole Sealing project, in particular Dr Nick Jefferies (Jacobs, UK). Thanks also to NWMO (Canada) and GTK (Finland) for in-kind contributions to the IBL project. Many thanks to KIC (Japan) for their permission to access the Tsukinuno mine and for their continued logistical support.

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9. Presentations

All presentations from the workshop are available on the NAWG website at https://www.natural-analogues.com/nawg-workshops/nawg-16-2019-japan.

10. Conclusions

The NAWG-16 workshop (October, 2019) in Zao, Yamagata, Japan should be seen as a success simply due to the determination of all who travelled from far and wide (both internationally and domestically) to get to the venue, despite the close attention of Taifun Hagibis.

The presentations themselves (see https://www.natural-analogues.com/nawg-workshops/nawg-16-2019-japan for details) covered a broad range of technical and socio-political themes but, when compared to earlier NAWG workshops (see https://www.natural-analogues.com/nawg-workshops/other-nawg-workshops for details), they were clearly much more focussed than has been the case in the past. Arguably, this reflects two things: first, the maturation of views on the use of NA information overall and, second, worldwide progress in developing repository concepts to the stage that studies such as NA can be applied in more specific supporting roles than ever before. Nevertheless, the wide range of focussed work presented here augers well for future NA studies and is a good example of the diversity of existing and planned work which stretches across process-specific studies (such as bentonite saturation) to efforts to offer clear information on issues of concern in radioactive waste disposal to as wide a range of stakeholders as possible.

Please note that it is currently foreseen that NAWG-17 will be held in Biel/Bienne (Switzerland) at the end of November, 2021 and NAWG-18 in Copenhagen at the end of August, 2022 (but please check www.natural-analogues.com for up-to-date details)[3].

[3] As noted before, this was not to be due to the COVID pandemic. At the time of writing, NAWG-17 will be held in May, 2023 (please see www.natural-analogues.com for details).

Acknowledgements

NAWG would like to thank the management and staff of Hotel Jirin (Zao Onsen, Japan) for their wonderful support throughout the period of the workshop. They went above and beyond the call of duty to ensure that all participants had a wonderful stay, it was greatly appreciated. Thanks to Dr Jon Engström (GTK) and Dr Ulrich Noseck (GRS) for their expertise in and support of the online portion of the workshop – they jumped in to help us out and didn’t let us down.

Thanks to NUMO (Tokyo, Japan), in particular to Mr M.Yamada, for their help throughout the event, from planning right through to successfully hosting the workshop. Thanks also to KIC (Tokyo. Japan), in particular to Dr M.Ito, for kindly allowing access to their sites.

Finally, many thanks to all the participants whose wholehearted engagement in all aspects of the meeting (both formal and informal) was a joy to behold.