Proceedings of the NAWG-17 Workshop

8-11th May, 2023, Zadar, Croatia

NAWG
NATURAL ANALOGUE
WORKING GROUP

University of Zagreb
FACULTY OF MINING,
GEOLOGY AND PETROLEUM
ENGINEERING

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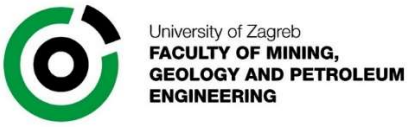
Workshop organized by:



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Contents

1. INTRODUCTION

Figure 1.1 Forum, Zadar, Croatia (by courtesy of Galla Uroić).

After a pause due to the COVID pandemic, the NAWG-17 workshop was held in the historical city of Zadar (Fig.1.1), Croatia (https://zadar.travel/) and was an opportunity to explore recent developments in the use of natural analogues in supporting safety cases for the disposal of radioactive waste.

Argumentation by use of analogy is well established in many fields including philosophy, biology, linguistics and law and most scientists are familiar with this approach and will have used it at some point in their career. The aim of workshop was to discuss:

Radioactive waste form stability: including vitrified waste stability and spent fuel evolution.

Repository near-field studies: including corrosion of copper and steel canisters and the long-term stability of the bentonite buffer.

Repository far-field studies: including self (or regional) analogues of potential repository sites and assessment of potential radionuclide releases to the host rock and biosphere

Stakeholder communication: including examples of approaches used to communicate different aspects of waste disposal to both technical and non-technical audiences

Workshop days with lively discussion on various uses of analogues were followed by a field trip to see the ancient Roman city of Colentum (the island of Murter) and its Roman concrete structures.

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1.1. WORKSHOP AGENDA

NAWG-17 Workshop

Monday, 8th to Thursday, 11th May, 2023, Hotel Kolovare, Zadar, Croatia

Monday 8th May		
	Travel to the venue	
17:00 – 18:00	Registration at the hotel pool	
18:00 – 20:00	Icebreaker and finger food at the swimming pool	
Tuesday 9th May		
	Introduction and registration	
09:00 – 09:10	Opening of the workshop and organisational aspects	Russell Alexander, NAWG chairman
09:10 – 09:20	Welcome to Zadar	Zelimir Veinovic, University of Zagreb
09:20 – 09:40	Current status of the Croatian national programme	Galla Uroic, Croatian Decommissioning Fund
Session I	**Exploring the current use of natural analogues in developing and supporting the safety case**	**Chair: Erika**
09:40 – 10:25	Natural Analogues – A proposed strategy for implementation within the Nuclear Waste Services (UK) programme of geological disposal	Simon Norris, NWS, UK
10:25 – 10:55	*Coffee/tea (with cakes)*	
10:55 – 11:25	A critically reviewed catalogue of NAs to support the Nuclear Waste Services (UK) programme of geological disposal	Russell Alexander, Bedrock Geosciences, Switzerland
11:25 – 11:45	Use of NAs in the Finnish safety case – the Complementary Considerations approach	Heini Reijonen, GTK, Finland
11:45 – 12:15	NAs and long-term evolution: upscaling towards repository relevant space and time scales	Venessa Montoya, SCK-CEN, Belgium
12:15 – 13:45	*Lunch*	

Session IIa	EBS – waste forms	**Chair: Simon**
13:45-14:15	Natural glass alteration under hyperalkaline conditions for about 4000 years	Ryosuke Kikuchi, University of Hokkaido, Japan
Session IIb	EBS - canisters	**Chair: Simon**
14:15 – 14:45	Multi-scale and isotopic analyses to understand the corrosion mechanisms of 400 year old archaeological iron analogues	Delphine Neff, CEA Saclay, France
14:45 – 15:15	MICA: overview of Phase I of the Michigan International Copper Analogue project	Ismo Aaltonen, GTK, Finland
15:15-15:45	A potential NA for the longevity of iron and steel in a clay from NW Scotland	Tony Milodowski, Independent, UK
15:45 – 16:15	*Coffee/tea (with cakes)*	
16:15 – 16:35	Archaeological analogues – an essential contribution to canister lifetime modelling	Vlatislav Kaspar, UJV Rez, Czech Republic
16:35 – 17:05	The use of NA information in assessing the chemical evolution of HLW disposal cells	Erika Neeft, COVRA, The Netherlands
17:05	Close of Day 1	
19:00	Dinner in the hotel	
Session III	**Posters: will be on display throughout the workshop**	
An application of research for buffer/backfill materials through estimation of Cs diffusion in compacted Taiwan bentonite		Chuan-Pin Lee, National Tsing Hua University, Taiwan
Bioreduction of aqueous uranium(VI) under conditions relevant for deep geological repository of nuclear waste		Dawoon Jeong, KAERI, South Korea
Advances in the understanding of radionuclides migration processes in natural analogues systems		Olga Riba, Amphos 21, Spain
Batch experiments for determining uranium dissolution kinetics using uranium-containing coaly		Hakyung Cho, Jeonbuk National University, South Korea

slate and groundwater from a natural analogue site in Korea	

Wednesday 10th May		
Session IIc	**EBS - bentonite**	**Chair: Andree**
09:00 – 09:30	The ongoing IBL project - recent results	Heini Reijonen, GTK, Finland
09:30 – 10:00	Microbiology of barrier component analogues of a deep geological repository	Rachel Beaver, University of Waterloo, Canada
10:00 – 10:30	Microbial occurrence and their growth in compacted bentonite in subsurface environments	Yuki Amano, JAEA, Japan
10:30 – 11:00	*Coffee/tea (with cakes)*	
11:00 – 11:30	Cigar Lake: behaviour of ^{129}I and implications for transport in clay barriers	Zhenze Li, CNSC, Canada
11:30 – 12:00	Fracture filling smectites as NAs for the bentonite buffer - case studies from Finland	Heini Reijonen, GTK, Finland
12:00 – 13:30	*Lunch*	
Session IIc (cont.)	**EBS - bentonite**	**Chair: Andree**
13:30-14:00	Thermal alteration of bentonite: an overview	Russell Alexander, Bedrock Geosciences, Switzerland
14:00-14:30	Experimental and geochemical modelling investigations of Cs, Sr and Co sorption on Zhisin Clay, Taiwan	Polly Tsai, INER, Taiwan
Session IVa	**Far-field studies - radionuclides in natural systems**	**Chair: Russell**
14:30 – 15:00	A natural analogue study in uranium deposits of the Okcheon Metamorphic Belt, Korea: basic investigations and future prospects	Min-Hoon Baik, KAERI, South Korea
15:00 – 15:30	*Coffee/tea (with cakes)*	
15:30 - 16:00	Water-rock interactions of uranium deposits: a field study in the Okchoen Metamorphic Belt, Korea and laboratory batch experiments	Sung-Wook Jeen, Jeonbuk National University, South Korea
Session IVb	**Far-field studies - climatic evolution**	**Chair: Russell**

16:00-16:30	NAs for long-term climato-tectonic evolution scenarios	Koen Beerten, SCK-CEN, Belgium
16:30	Close of Day 2	
	Free evening – enjoy Zadar	
Thursday 11th May		
Session IVc	**Far-field studies - rock matrix diffusion**	**Chair: Polly**
09:00-09:40	Analogue Studies of Rock Matrix Diffusion in Higher Strength Rocks	Simon Norris, NWS, UK
09:40-10:20	Matrix diffusion at the small scale and the role of heterogeneity	Marja Siitari-Kauppi, University of Helsinki, Finland
Session VI	**General studies**	**Chair: Stephane**
10:20 – 10:50	Ultra-trace analysis with AMS in the frame of experiments relevant for nuclear waste disposal	Francesca Quinto, INE-KIT, Germany
10:50-11:20	*Coffee/tea (with cakes)*	
11:20 – 11:50	Recent activities on NA studies in Japan	Satoru Suzuki, NUMO, Japan
Session VII	**Excursion**	**Chair: Stephane**
11:50 – 12:10	Concrete longevity studies: current status and future potential	Russell Alexander, Bedrock Geosciences, Switzerland
12:10 – 12:30	Introduction to the excursion sites	Jagor Blazic, Independent, Croatia
12:30	Close of the workshop	Russell
12:45	Leave from the front of the hotel by bus	Packed lunch will be provided on the bus
ca. 18:00	Return to the hotel	
19:30	Workshop dinner in a local restaurant	

1.2. FIELDTRIP

The field trip was led by Mr. Jagor Blažić, B.Sc.Archeol. expert in the field and organized as a visit to the "*Colentum*" (Fig. 1.2) location on the island of Murter. The city of Colentum was a Liburnian and later Roman city. It was founded by Liburnians in the 11th century, and has kept the continuity of population until the present day.

Colentum, along with Iader, was the most important port of southern Liburnia, and its existence was based on agriculture, farming, seafaring and fishing. It became a Roman town in 1st century BC and the remains have features of typical Roman architecture – houses, cisterns, baths and streets.

Figure 1.2 Remains of the Roman city of Colentum, Murter, Croatia.

Roman Maritime Villa (Fig. 1.3 & 4) was built in 1st century AD, next to the sea (sea level rose by 1.6 meters). It was a large building, 100 x 50 meters, constructed in several phases with porch, 10 coastal rooms, courtyard, cistern and baths, decorated with marble tiles, mosaics and frescos. Many fragments of ceramics and roof tiles were found as well. It was a multi-storied buildings, probably demolished in an earthquake (archaeological indicators). It consisted of vertically inserted poles and pillars, and horizontally laid beams. In the vicinity, large amounts of amphora corks, and various fragments of amphorae, ceramic ware and an almost entirely preserved ceramic oil lantern were discovered.

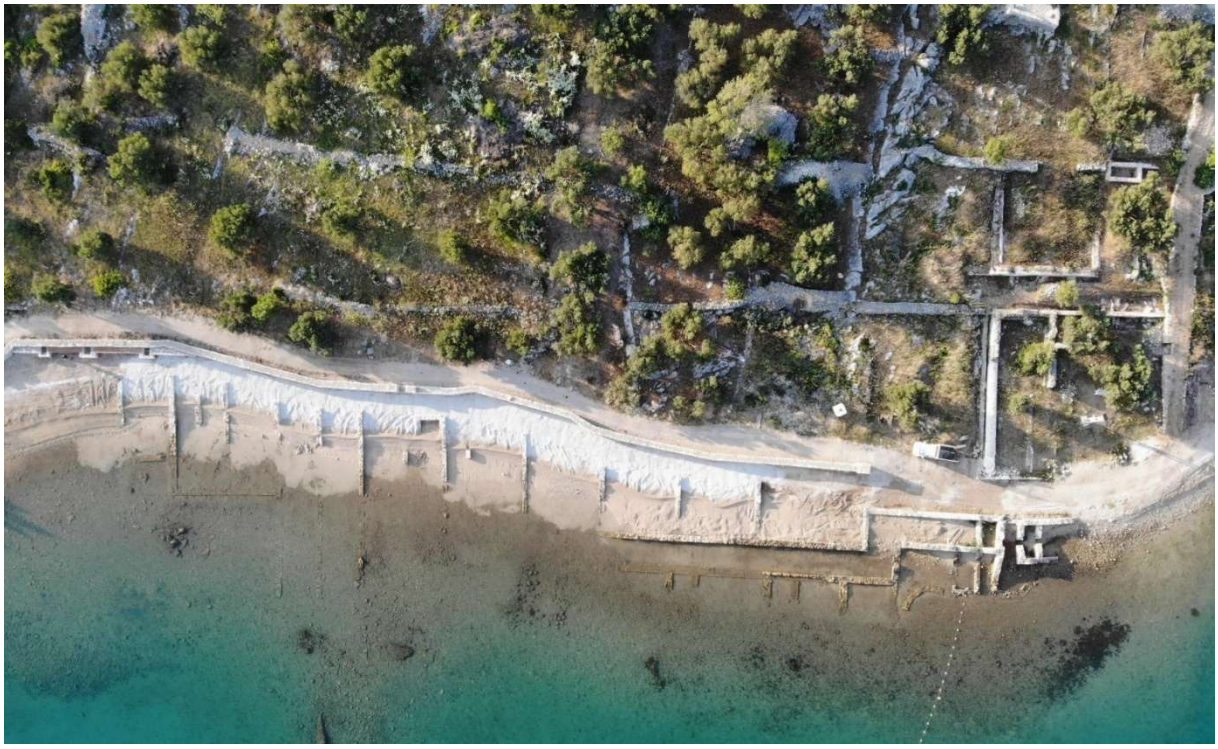

Figure 1.3 Remains of Roman Maritime Villa, aerial photo.

Figure 1.4 Remains of Roman Maritime Villa.

Roman Cistern (Fig. 1.5), called “the Dungeon” because of its appearance, was filled with rainwater and belonged to a complex of baths. It was built with Roman concrete (*opus caementicium*) – durable building material/technology, and the interior of the cistern was lined with hydraulic mortar – watertight material used in antiquity.

Figure 1.5 Remains of Roman cistern.

Opus Caementicium (Fig. 1.6) technology was in widespread use (from 2nd century BC) for construction in ancient Rome, since its strength was enhanced by addition of pozzolanic ash and it had self-repairing properties. It was able to set underwater. Vitruvius (c. 80–70 BC – after c. 15 BC), Roman architect and engineer, recommended pozzolana, volcanic ash, as an additive for structural lime mortar, with aggregates varied from rock, ceramics and lime clasts to brick rubble. Pozzolana concrete was more resistant to salt water than modern-day concrete. Since lime clasts react with water, new calcium carbonate crystals formed and reseal the cracks. Roman concrete, containing volcanic ash and quicklime, benefited form reacting with seawater creating tobermorite, a rare crystal which may resist fracturing. It is certainly a candidate for the most durable building material in human history.

Figure 1.5 Remains of Roman concrete (opus caementicium).

1.3. LIST OF PARTICIPANTS

Participants name	Organisation & country
Andree Lommerzheim	BGE Technology, Germany
Milena Schönhofen-Romer	BGE, Germany
Bastian Graupner	ENSI, Switzerland
Calum Powrie	NWS, UK
Chuan-Pin Lee	National Tsing Hua University, ROC
Dawoon Jeong	KAERI, ROK
Delphine Neff	CEA, France
Erika Neeft	COVRA, Netherlands
Francesca Quinto	INE-KIT, Germany
Galla Uroic	University of Zagreb, Croatia
Hakyung Cho	Jeonbuk National University, ROK
Heini Reijonen	GTK, Finland
Hsieh Hsien Te	NARI, ROC
Ismo Aaltonen	GTK, Finland
Jagor Blazic	Independent, Croatia
Kanya Kimura	Hokkaido University, Japan
Koen Beerten	SCK-CEN, Belgium
Marja Siitari-Kauppi	University of Helsinki, Finland
Min-Hoon Baik	KAERI, ROK
Mostafa Fayek	University of Manitoba, Canada
Olga Riba	Amphos 21, Spain
Polly Tsai	NARI, ROC
Rachel Beaver	University of Waterloo, Canada
Russell Alexander	Bedrock Geosciences, Switzerland
Ryosuke Kikuchi	Hokkaido University, Japan
Satoru Suzuki	NUMO, Japan
Simon Norris	NWS, UK
Stephane Brassinnes	ONDRAF/NIRAS, Belgium
Sung-Wook Jeen	Jeonbuk National University, ROK
Tony Milodowski	Independent, UK
Vanessa Montoya	SCK-CEN, Belgium
Vlastislav Kaspar	UJV Rez, Czech Republic
Yuki Amano	JAEA, Japan
Zhenze Li	CNSC, Canada

Figure 1.6. Participants of the NAWG-17 Workshop.

1.4. 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 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 exposure (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).

1.5. Introduction to natural analogues – what are they and how do they help us assess repository 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 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, 2023; Reijonen & Alexander, 2024, 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). 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.

2. ABSTRACTS

Figure 2.1. Remains of Roman Maritime Villa, Murter, Croatia.

Multiscale and isotopic analyses to understand the corrosion mechanisms of 400 years archaeological iron analogues

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Abstract

The corrosion of steel in anoxic carbonated media over centuries is a crucial issue for the preservation of archaeological artefacts or the prediction of long term corrosion of metallic elements in a nuclear waste deep geological repository. Understanding the mechanisms controlling the corrosion kinetics is necessary and archaeological analogues are one of the pillar for such studies. Specifically, in laboratory simulation as well as on archaeological analogues, in addition to a layer of several 100 µm made of iron carbonates (siderite, chukanovite), a submicrometric layer was detected at the metal/corrosion product interface that questioned the corrosion processes.

This talk presents the methodology developed on 400 year archaeological analogues to determine the physico-chemical properties of such interfacial nanolayer. Transverse sections and thin films (FIB) were studied by a multitechnique approach determining the phases' morphology, distribution and nature at micrometric and nanometric scales (µraman spectroscopy, TEM, STXM under synchrotron). A continuous layer made of a mix of FeII and FeIII species (maghemite/magnetite) was identified at the metal/CPL interface. In addition, the investigated archaeological artefacts were put in a D_2O desaerated solution, during several months. NanoSIMS observation of D/H ratio distribution showed the interfacial layer was less porous than the carbonate layer. Last, local conductivity was determined inside the corrosion layer by C-AFM. Nanometric conductive pathways were identified through the insulating carbonate layer. This methodology allowed to refine the understanding of the corrosion mechanisms from the viewpoint of long term prediction and will be applied to copper archaeological analogues.

Keywords: archaeological analogues, corrosion, steel, multiscale characterisation

A critically reviewed catalogue of NAs to support the Nuclear Waste Services (UK) programme of geological disposal

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Abstract

The successful implementation of a repository for radioactive waste in the UK will require an assessment of the ability of the facility to meet appropriate regulatory safety criteria at all stages during its construction, operation and post-closure. As part of this, an environmental safety case (ESC) will be presented which will contain a collection of claims, arguments and evidence (CAE) which collectively demonstrate that long-term safety can be achieved and maintained. Natural analogues (NA) can be helpful in demonstrating understanding of aspects of repository performance by, e.g., providing evidence that certain materials can survive for long periods.

Appropriate NAs can be critical to providing long-term practical demonstrations to support the theoretical and mathematical arguments of the ESC and they may have a significant role in the overall process understanding. As part of the ongoing repository programme in the UK, an updated version of an existing catalogue of NA studies (focussed on the requirements of the ESC) has been produced and the background to the catalogue is presented here.

During the update, relevance screening was carried out, meaning that some information has been substituted as it was felt to be less relevant to the current and foreseen requirements of the UK national programme. Component level gap analysis was also performed, leading to additions at the topical level. In addition, the IFEP database has been mapped to the contents of the catalogue as a first step towards full integration of the NA Catalogue in the overall repository programme knowledge base.

Keywords: natural analogues; geological disposal facility; long-term behaviour; long-term processes; large-scale processes

Integrated assessment of thermal alteration of bentonite

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Abstract

In the case of thermal alteration of bentonite, various changes (e.g. illitisation, cementation, etc. – see Reijonen et al., 2023 for an overview) can occur. Currently, in many national programmes, the requirements for maximum bentonite temperatures are set around 100°C. In many cases, there would be interest in increasing this limit, so allowing, for example, the placement of waste packages more closely than is currently planned. But this would require more stringent justification than currently exists.

Although there are significant number of NA papers on this theme in the literature, a recent re-analysis (Posiva, 2013, 2023) shows that most examples suffer from the same problem: the conditions are not truly representative of a repository. In most cases of contact metamorphism studied, the temperatures have been much too high (800-900°C rather than the repository relevant maximum of 100°C). In addition, the degree of saturation of the EBS affects the thermal conductivity of the bentonite (the highest temperatures being met in the "dry" case, i.e. bentonite water contents being as installed), but the saturation state of the bentonite has not been examined in any thermal NA studies.

Here, a novel two-pronged approach in proposed where information from well-controlled URL experiments (e.g. FEBEX, Lanyon & Gaus, 2013; EURAD-HITEC, Villar et al., 2020) is data-mined and coupled with a focussed NA study which will look at a system under repository-relevant environmental conditions, relevant temperature fields and will assess the full physico-chemical conditions of the bentonite (including saturation states) using modern analytical methods.

Keywords: natural analogues; thermal alteration; bentonite cementation; integrated approach; radioactive waste disposal

References

Lanyon G.W., & Gaus I. (Eds.) (2013). Main outcomes and review of the FEBEX In Situ Test (GTS) and Mock-Up after 15 years of operation. Nagra Arbeitsbericht. NAB 15-04. Nagra, Wettingen, Switzerland.

Posiva (2013). Safety case for the disposal of spent nuclear fuel at Olkiluoto – Complementary Considerations 2012. Posiva Report 2012-11, Posiva, Eurajoki, Finland.

Posiva (2023). Safety Case for the Operating Licence Application - Complementary Considerations (CC). Posiva Report 2021-02. Posiva, Eurajoki, Finland.

Reijonen, H., Elminen, T., Heikkilä, P., Kuva, J., Jolis, E. 2023. Enhanced identification of fracture smectites and other alteration minerals via short wave infrared reflectance at two Finnish crystalline sites, Olkiluoto and Hyrkkölä. Rock Mechanics and Rock Engineering (submitted).

Villar, M.V., Armand, G., Conil, N., de Lesquen, Ch., Herold, Ph., Simo, E., Mayor, J.C., Dizier, A., Li, X., Chen, G., Leupin, O., Niskanen, M., Bailey, M., Thompson, S., Svensson, D., Sellin, P., Hausmannova, L.(2020). D7.1 HITEC. Initial State-of-the-Art on THM behaviour of i) Buffer clay materials and of ii) Host clay materials. Deliverable D7.1 HITEC. EURAD Project, Horizon 2020 No.847593. 214 pp

Long-term durability of concrete

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Abstract

National radioactive waste disposal regulators have been calling for a less conservative approach to the assessment of engineered barrier degradation: for example, in 2009, the Swedish regulator, SSM, noted that it would like to see the long-term degradation of cementitious materials (waste, containers, backfill, tunnel liners) treated in a more realistic manner.

Current treatment is highly simplistic in most national programmes, utilising simple mixing tank approaches with emphasis on over-prediction of consequences (i.e. relatively rapid degradation of the cement leading to release of radionuclides to the surrounding host rock). Even when more sophisticated reactive transport codes are used for these assessments, they are generally supported by only short-term laboratory experiments, so it is perhaps not surprising that longer-term processes are treated in an over-conservative manner (due to a lack of relevant data).

Here, relevant natural and archaeological analogues to assess the long-term degradation of both OPC (Ordinary Portland Cement) and low-alkali (or low heat) cements are proposed as is an experimental approach to provide relevant data for the modelling of long-term concrete durability.

Keywords: radioactive waste; concrete degradation; natural analogues; archaeological analogues; degradation modelling

Fracture filling smectites as NAs for the bentonite buffer - case studies from Finland

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Abstract

Fracture filling minerals have been recently studied at four different crystalline sites, Olkiluoto, Hyrkkölä, Romuvaara and Kivetty, in Finland with the aim of understanding the smectite mode of occurrence in a range of geological environments. The sites differ regarding the overall rock type (migmatites to granitoids) and current deep groundwater chemistry (saline to fresh). In all of the sites studied, several types of smectites have been identified, including montmorillonite, the mineral comprising the bulk of the bentonite planned to be used in the geological repositories for high-level radioactive waste. In some cases, even Na and Ca variants of montmorillonite have been identified. Detailed pertrographic examination using scanning electron microscopy show that smectites are formed at the late stage of ancient hydrothermal alteration and clear replacement textures are observed, providing evidence of the fracture filling smectites are preserved in situ, even in open fracture systems. Results confirm the previous smectite observations within depth range down to 1 km depth, and further defines the smectite mineralogy. The volumes of the fracture filling minerals is very small compared to bentonite buffer used in the repository designs, but they provide a good basis to examine the overall stability analogue in the relevant environments. In this presentation, an overview of the research is made, and some preliminary results are discussed.

Keywords: ”fracture filling mineral”, “bentonite”, ”smectite”, “montmorillonite”, “faults”

The ongoing IBL project - recent results

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Abstract

International Bentonite Longevity (IBL) project has recently produced relevant background information to support further work in ongoing Phase B. Multimodal analysis results have been obtained from drillhole D1 (bed #19) sampled during Phase A, including cation exchange capacity, exchangeable cation composition, quantitative mineralogy and geochemistry. In this presentation we present the combined results and use them to validate hyperspectral analysis of the same core. Within a relatively thin bentonite bed, < 0.5 m, significant variation in various parameters are observed, specifically around the contacts to under and overlying mud rocks. The results are in agreement with the textural observations made via X-ray tomography. They are of specific importance, as to date, detailed descriptions of variations within single bentonite beds has been very scarce in the literature. The results showcase the importance of detailed investigations in order to assess the relevance of the analogy of natural bentonites to repository bentonites in respect to homogeneity and overall mineralogy of the material being studied. The results obtained here can be used to aid in the future analytical programmes in the IBL-project, but can also be considered for any bentonite deposit characterisation.

Keywords: ”geochemistry”, “bentonite”, ”smectite”, “montmorillonite”, “hyperspectral analysis”

Acknowledgements

IBL-project organisations from Phase A (NWS, UK; Jacobs, UK) and B (NWMO, Canada; NUMO, Japan, Bedrock Geosciences, Switzerland; GTK, Finland) are acknowledged for making this research possible. Associated laboratories are mentioned in the presentation. Special thanks goes to Kunimine Industries (KIC) and M. Ito, S. Norris (NWS), W.R. Alexander (Bedrock Geosciences), E. Kremer (NWMO) and M. Yamada (NUMO).

Use of NAs in the Finnish safety case – the Complementary Considerations approach

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Abstract

Posiva has published the main reports of the safety case in a digital safety case format (cms.posiva.fi) of which Complementary Considerations is the main report discussing natural analogues (NA) in the safety case. In addition to NA, the report discusses the potential hazard arising from SNF and LILW repositories in context by comparing the radiotoxicity indexes to natural occurrences of radioactive media. It also briefly discusses the justification of geological disposal in general. NAs are discussed in a site and design specific manner, reflecting the needs of operational licence application for the disposal facility. The contents of the report regarding NA is divided in broad categories of waste, container materials, clay materials, host rock and external processes. The content of the report is connected to the screening results of relevant features, events and processes (FEPs) as well as final safety statements made in the safety case. This presentation provides an overview of the approach taken and some examples of the main argumentation arising from the NA knowledgebase.

Keywords: “geological disposal”, “natural analogues”, “complementary considerations”, “safety case”, “long-term safety”

Experimental and geochemical modelling investigations of Cs, Sr and Co sorption on Zhisin Clay, Taiwan

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Abstract

This paper aims to compare the results between batch sorption tests and geochemical modelling for the sorption of key nuclides (Cs-137, Sr-90 and Co-60) on Zhisin soil, a natural clay material located in in Chang Yuan Village, Taitung County, the Coastal Range of eastern Taiwan. The clay has been identified as a Ca-bentonite and the main minerals are montmorillonite (~35%) and kaolinite (~28%) using XRD quantitative analysis. The geochemical model was performed with PHREEQC and several thermodynamic database for homogenous aqueous oxidative/ reductive reactions, surface complex reactions and cation exchange reactions were used. The results showed that the Kd value obtained from model prediction for Cs on the clay in the synthetic groundwater (SGW) was in agreement with that of sorption experiment. The Kd values of Sr and Co were also within one order magnitude between experimental results and model prediction.

Although Zhsin clay has good adsorption capacity for radionuclides, the mechanical and thermal stabilities 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 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 resulted in cost in-effectiveness for mining. However, it can be considered as an evidence of natural analogue study for local bentonite.

Keywords: Zhisin Clay; Geochemical Modelling; Batch Sorption Test; Distribution Coefficient (Kd); Thermodynamic Database

Bioreduction of aqueous uranium(VI) under conditions relevant for deep geological repository of nuclear waste

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Abstract

This study was performed to assess the potential of U(VI) reduction by indigenous bacteria under conditions relevant for deep geological repository (DGR). Three different indigenous bacteria were obtained from granitic groundwater at depths of 44–60 m (S1), 92–116 m (S2), and 234–244 m (S3). The S2 groundwater contained the highest U concentration of 885.4 μg/L among three groundwater samples, where U mainly existed in the form of Ca2UO2(CO3)3(aq). The S2 groundwater amended 20 mM of sodium acetate as an electron donor was used for the aqueous U(VI) ((U(VI)aq) bioreduction experiment. The U(VI)aq bioreduction experiments were conducted under anaerobic conditions for 24 weeks. Variations in the U(VI)aq concentration were monitored for 24 weeks to compare U(VI)aq removal efficiency in response to indigenous bacteria. The highest U(VI)aq removal efficiency of 57.8% was observed in S3, followed by S2 (43.1%) and S1 (37.7%). The incomplete U(VI)aq removal was attributed to the presence of the thermodynamically stable uranyl carbonate complex. Changes in indigenous bacterial communities were observed by a high-throughput 16S rRNA gene sequencing analysis. After 24 weeks of anaerobic reaction, sulfate-reducing bacteria populations including Thermodesulfovibrio yellowstonii and Desulfatirhabdium butyrativorans were significantly increased in the S3 sample, which contributed to the highest U(VI)aq removal efficiency of 57.8%. The precipitates produced by bacterial activity were determined to be U(IV)-silicate nanoparticles by a transmission electron microscope (TEM)-energy dispersive spectroscope (EDS) analysis. These results demonstrated that considerable U immobilization is possible by stimulating the activity of indigenous bacteria in the DGR environment.

Keywords: Uranium, Bioreduction, Indigenous bacteria, Illumina Miseq analysis, Sulfate-reducing bacteria

A Natural Analogue Study in Uranium Deposits of the Ogcheon Metamorphic Belt: Basic Investigations and Future Prospect

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Abstract

We introduce a research project of natural analogue study which is being carried out as a part of national R&D programme for storage and disposal of spent nuclear fuel in Korea. In addition, we introduce some results for long-term geochemical behaviours of uranium in a granitic rock-groundwater system which has been carried out in the underground research facility, KAERI underground Research Tunnel (KURT). A uranium deposit in the Ogcheon Metamorphic Belt (OMB) was selected as a natural analogue study site to investigate long-term radionuclide behaviour in high-level radioactive waste repository environments. Various basic investigations for groundwater and rock samples from the study site were investigated and then geochemical behaviour of uranium in the groundwater and rock were studied. Results show that U(IV)-minerals such as uraninite and uranothorite have been preserved without transformation to U(VI)-minerals and that U leaching/dissolution from rock into groundwater has been limited. This may be due to a well-maintained redox condition and a restricted contact with oxidizing groundwater. In the future, reasons and mechanisms of the results will be further investigated and few more study sites in the OMB will be developed and investigated for natural analogue studies of radionuclide migration and retardation.

Keywords: Natural analogue study; Uranium deposit; Ogcheon metamorphic belt; Uranium species; Uranium mineral; Uranium mobility

An application research for buffer/backfill materials through estimation of Cs diffusion in compacted Taiwan bentonite

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Abstract

The migration of HTO and Cs in Taiwan bentonite(Zhisin) clay was studied by traditional through-diffusion (TD) experiments and a non-equilibrium analysis to determine diffusion coefficients and retardation factor. A numerical analysis with a minimum error for the HTO and Cs diffusion coefficients in compacted Zhisin clay was conducted. The TD experimental results and numerical analysis showed that diffusion of HTO and Cs reached steady state within 10 and 120 days, respectively, and diffusion coefficients decreased with the increases in the compacted density. In fact, there was retardation of Cs diffusion in Zhisin clay. A two-site sorption model for Cs in Zhisin clay was applied to simulate fast and slow sorption behavior quantitatively.

Keywords: caesium, HTO, bentonite, nonlinear , adsorption

Advances in understanding radionuclides migration processes in natural analogues systems

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Abstract

The process understanding and quantification of natural, archaeological and industrial sites, i.e. Natural Analogues, can provide with insights on the long-term processes and physical - chemical behaviour of engineered or geological barrier systems over very long timescales relevant to post-closure safety of disposal facilities.

Scientific literature review of natural analogues studies published since 2010 have been performed in the context of assessing the migration/retention processes of radionuclides relevant for repository systems considered in Belgium for high-level radioactive wastes. The main migration /retention processes of the radionuclides have been identified for the different studied sites:

i) Solubility of primary phases: Oman, Ruprechtov

ii) Retention in primary phases or in secondary phases by precipitation and co-precipitation: Maqarin, Oman, Oklo, Cigar Lake, Ruprechtov, Tono, Daejeon, El Berrocal, Mina Fe, Palmottu, Krunkelbach Valley, Coles Hill, Nopal I, Poços de Caldas, Koongarra, Streltsovka caldera, Rosemanowes Quarry.

iii) Adsorption on minerals surfaces / colloids: Oklo and Bangombé, El Berrocal, Palmottu, Idaho National Laboratory, Coles Hill, Nopal I, Koongarra, Streltsovka caldera.

iv) Retention in colloids or particulate matter: Neddle's Eye, Poços de Caldas

v) Diffusion through matrix: Palmottu, Poços de Caldas

vi) Complexation on organic matter: Neddle's Eye

Based on the outcomes of this review, and in accordance with physical and geochemical characteristics of each site, different key chemical processes have been identified driving the retention of radionuclides in the studied systems. This information might be used to provide with safety relevant drivers for the design of the engineered barrier system and supporting documents for safety cases.

Keywords: nuclear waste repository; radionuclides; migration; natural analogues.

The use of analogue information in assessing the chemical evolution of HLW disposal cells

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Abstract

The broader scope of the EURAD work package ACED (Assessment of Chemical Evolution of ILW (Intermediate Level Waste) and HLW (High Level Waste) Disposal cells) is the assessment of the chemical evolution at the disposal cell scale, considering disposal concepts that are representative for European disposal concepts. HLW and ILW disposal cells in granitic and clay host rocks are studied in ACED. The disposal concept for vitrified HLW in these assessments comprises the encapsulation of this processed waste in a carbon steel overpack and surrounding this overpack with a concrete or bentonite buffer. The radionuclide mainly responsible for the radiation dose rate at the overpack/buffer interface is 137mBa, a daughter of 137Cs. The combination of the guaranteed maximum content of this radionuclide in vitrified HLW, its homogeneous distribution within the vitrified waste form, the cooling time envisaged in European disposal concepts and the shielding provided by steel and glass generates a radiation dose rate that is smaller than the minimum in experimental dose rates showing radiation enhanced steel corrosion. The exclusion of radiation enhanced corrosion facilitates the use of natural analogues of iron interfacing clays and/or archaeological analogues of reinforced concrete for the identification of relevant processes for assessing the chemical evolution of disposal cells. The available analogues in literature for HLW disposal cells are shown.

Keywords: vitrified waste; radiation; steel; concrete; bentonite

Batch experiments for determining uranium dissolution kinetics using uranium-containing coaly slate and groundwater from a natural analogue site in Korea

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Abstract

This study evaluated the water-rock interactions, in which uranium is involved in a particular groundwater system. Batch experiments were conducted under various geochemical conditions resulting from water-rock interactions containing uranium. The batch experiments used artificial groundwater simulating the groundwater of Boeun-gun (county) and coaly slate containing uranium from the Okcheon Metamorphic Belt, a natural analogue study site in Korea. The artificial groundwater was prepared with different pH conditions (i.e., initial pHs of 5, 7.9, and 9). In addition, uranium (2 mg L-1) was spiked in another batch experiment. The main constituent minerals of the coaly slate were quartz, pyrite, sphalerite, coal materials, and uranium minerals (e.g., uraninite and ekanite). The pH of all batch experiments decreased rapidly after the start of the reactions. It may be due to the absence of carbonate and the generation of hydrogen ion by oxidation of iron sulfide. The concentrations of uranium in the initial pH 7.9 experiment increased due to dissolution of uraninite and ekanite. In the experiment where uranium was spiked, the uranium concentration decreased over time, indicating that it could be controlled by adsorption or surface complexation with iron (hydr)oxide. In all batch experiments, aqueous complexes of uranium existed as UO_2SO_4(aq) after the reactions. Thus, uranium in groundwater formed complexes with sulfate, and its mobility could be limited by adsorption or surface complexation with iron (hydr)oxide.

Keywords: Batch experiments, uranium, iron (hydr)oxide, adsorption, surface complex

Water-rock interactions of uranium deposits: a field study in the Okcheon Metamorphic Belt, Korea and laboratory batch experiments

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Abstract

Geochemical behaviors of uranium in the far-field of the high-level radioactive waste deep geological repository are significantly influenced by the interactions between rock and groundwater. Natural analogue studies are required to understand the long-term behaviors of uranium in hydrogeological systems in the geological time scale. In this regard, to understand the mobility of uranium in groundwater, it is necessary to evaluate the water-rock interactions in a particular site. This study evaluated the hydrogeochemical characteristics of groundwater, mine water, and quarry runoff in the Okcheon Metamorphic Belt, a natural analogue study site in Korea, containing uranium ore bodies. In addition, batch experiments were performed to evaluate the mobility of uranium resulting from the water-rock interactions. The study area is located at Boeun-gun (country) and Geumsan-gun (county) in the Okcheon Metamorphic Belt. In Boeun-gun, groundwater was sampled from four wells installed along the uranium ore bodies. In Geumsan-gun, mine water from the Birey deposit and runoff water from a quarry were sampled. Water quality measurements and ion analysis were conducted to investigate the geochemical conditions. The elemental composition of coaly slate containing uranium ore bodies, collected near the groundwater wells, was also analyzed, and the same analysis was performed for the sediments from the mine and quarry. The batch experiments were conducted for 14 days using the coaly slate and artificial groundwater that simulated groundwater in Boeun-gun (dissolved oxygen (DO) was below 1 mg L-1). The results of the field study showed that groundwater was under the slightly reduced condition with a relatively low Eh value (104.9 mV). The uranium in groundwater was shown to form aqueous complexes with carbonate (i.e., $UO_2(CO_3)_3^{4-}$ and $UO_2(CO_3)_2^{2-}$), and the mobility of uranium could be limited by co-precipitation with iron (hydr)oxides. The main component of the sediment in the mine was iron (hydr)oxide, an oxidation product of iron sulfide, and it contained a relatively large amount of uranium. It showed that minerals such as ferrihydrite ($Fe(OH)_3$) and hematite (Fe_2O_3) could co-precipitate uranium, limiting its mobility. Surface water, mine water, and quarry runoff were all under oxidized conditions and formed complexes with carbonates (i.e., UO_2CO_3, $UO_2(CO_3)_2^{2-}$, and $UO_2(CO_3)_3^{4-}$). The results of the batch experiment showed that the change in uranium concentrations with time increased at the beginning of the reaction, but decreased after 5 days. The increased concentrations of uranium seemed to be a result of the dissolution of the uranium minerals such as uraninite and ekanite. The decreased concentrations of uranium were considered to be a result of adsorption or surface complexation with iron (hydr)oxide, such as ferrihydrite, and SiO_2. The aqueous complex of uranium existed as a uranyl hydroxide complex ($(UO2)_3(OH)_7^-$) before the reaction and then formed a complex with sulfate ($UO_2SO_4(aq)$) after the reaction. Therefore, in the Okcheon Metamorphic Belt, uranium in groundwater formed complexes with carbonate and sulfate, but its mobility could be limited by co-precipitation or surface complexation with iron(hydro)oxide.

Keywords: Uranium, Okcheon Metamorphic Belt, iron (hydr)oxide, redox condition, geochemical behavior

Acknowledgements

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Ultra-trace analysis with AMS in the frame of experiments relevant for nuclear waste disposal

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Abstract

At the Grimsel Test Site (GTS), NAGRA´s generic underground research laboratory, radionuclide migration in near natural and repository relevant conditions is studied with several in-situ radionuclide tracer tests. With the CFM Long-term In-situ Test (LIT) we have investigated the near-field release of the radionuclide tracers 99Tc, 233U, 237Np, 242Pu and 241Am through compacted bentonite and a water conductive shear zone in the granodiorite rock during 4.5 years. Furthermore, we have designed to explore global fallout derived 236U, 237Np and 239Pu in groundwater samples of the GTS to try and determine their possible downward migration over a period of ca. 60 years and through the 450 m massive thrust sheet of crystalline rock. Such experiments are characterized by ultra-trace levels of radionuclides requiring the use of a highly sensitive analytical technique, like AMS that provides detection efficiency for actinides and 99Tc of ca. 1 × 104 atoms (25 ag) and 3 × 106 atoms (0.5 fg) in a sample, respectively [1, 2]. Results from LIT suggest that the radionuclide tracers were strongly captured by the bentonite with only trace releases of 99Tc, 233U, 237Np and 241Am during the saturation phase of the bentonite and afterwards a steady state release of 99Tc pointing to diffusive transport of a small fraction of not reduced TcO4-. Preliminary results from the study on global fallout actinides are of uncertain interpretation, but they constitute a proof of concept for further investigations at the GTS as well as at other natural analogues systems.

Keywords: in-situ radionuclide tracer tests; global fallout actinides; compacted bentonite; granodiorite rock; AMS.

References

[1] P. Steier et al., Nucl. Instrum. Methods Phys. Res., Sect. B, 268, 1045 (2010).
[2] F. Quinto et al., Anal. Chem., 91, 4585 (2019)

Matrix diffusion at the small scale and the role of heterogeneity

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Abstract

The existence and the extent of matrix diffusion, in fractured crystalline rock, has recently been discussed for its use in the safety assessments of spent nuclear fuel disposal. In this paper, we present how matrix diffusion has been studied in a variety of heterogeneous crystalline rock samples at the laboratory scale. The spatial distribution of porosity have been determined by C-14 PMMA autoradiography and these have been combined with the mineral structures, and the data are used to build up a 3D pore structure of rock samples with the aid of CT tomography for interpreting the results from different diffusion experiments. In addition, we present a multidisciplinary study on a U-rich muscovite granite boulder sample that gives insight to explain the presence of uranium and the extent of matrix diffusion, thus justifying the use of matrix diffusion in safety assessments. Multidisciplinary studies included pore structure characterization, mineralogical studies, observation of uranium phases, uranium series equilibrium (USD) studies, and Fe^{3+} analyses. The permeability and effective diffusion coefficient in the studied samples were determined using laboratory scale through-diffusion experiments with different probe molecules.

The results indicate that matrix diffusion is an important phenomenon and its extent depends on the connected pore structure of the host rock as well as on the mineral composition.

Keywords: Matrix diffusion, porosity, crystalline rock, heterogeneity

Microbial occurrence and their growth in compacted bentonite in subsurface environments

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Abstract

High-level radioactive wastes (HLW) from nuclear energy production must be disposed safely for at least tens of thousands years. The safety requirements for HLW 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. To evaluate long-term behaviour of microbial activity in bentonite buffer materials in subsurface environments, we have conducted an engineering scale of in-situ corrosion experiment at the Horonobe Underground Research Laboratory (URL) and field investigation of ancient bentonite rocks at the Tsukinuno bentonite deposit in Japan.

The in-situ experiment was set up and operated in sedimentary rocks at 350 m gallery of the Horonobe URL, and the bentonite samples were collected for microbial analyses after 3.5-year experimental duration. The bentonite rocks samples were collected from the tunnel wall in the Tsukinuno mine. The results of these microbiological investigations indicated that microbial activity could have been suppressed for a long period of time under high dry density conditions, although microorganisms could grow in the buffer material when the condition of pore size in buffer material would be heterogeneous. It is important that pore size distribution in buffer materials would be homogeneity.

Keywords: “Microbiologically influenced corrosion”, “Horonobe Underground Research Laboratory”, “bentonite buffer materials”, “Tsukinuno bentonite mine”

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Archaeological analogues – an essential contribution to canister lifetime modelling

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Abstract

More than 200 artifacts with the age of 500-2600 years have been picked up from 15 different archaeological sites in the Czech Republic. Adjacent soils have been examined – they were coarser than bentonites and lacked swelling ability. Pore solutions of the soils were similar to bentonite ones; they differed in cation composition, but anion composition, which is more important for corrosion behaviour, was similar, as demonstrated compared to Czech BCV bentonite. The environment was never completely anaerobic, main compounds were oxides and oxyhydroxides. Carbonates based corrosion products, requiring very low oxidation-reduction potential, were not detected unlike in previous lab and in-situ experiments. Nevertheless, the oxygen transport was very slow, and contribution of aerobic corrosion was negligible compared to anaerobic corrosion. Geochemical modelling was performed for selected sites based on the pore solutions composition.

The artifacts have been subjected to XRD analysis. It was possible to gain corrosion products from some. Some of the artifacts with low historical value were used for studying crosscuts. The evaluation of the artifacts revealed an important factor impacting the corrosion mechanism, which manifests only after sufficiently long period of being buried in the soil. While in the early stage the pore system is gradually being filled with corrosion products, thus reducing the corrosion rate, in latter stage the transport of iron cations is reduced too much, and newly formed corrosion products cause mechanical damage to previously formed ones. This phenomenon must be considered for a correct canister lifetime estimation.

Keywords: archaeological analogues; corrosion products; radioactive waste canister lifetime; deep geological repository; BCV bentonite

A potential Natural Analogue for the longevity of iron and steel in a clayey matrix from northwest Scotland, U.K.

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Abstract

Many repository designs for the geological disposal of radioactive wastes and spent nuclear fuel envisage placing the waste form in durable metal containers made of cast iron or steel (in some cases in combination with a more corrosion-resistant copper over-pack), as part of an engineered barrier system. The container serves to: (i) provide appropriate shielding during waste package handing; (ii) provide structural integrity to the waste package, and; (iii) to protect the waste form from groundwater penetration and subsequent leaching of radionuclides. Design requirements for the container to resist degradation and penetration by groundwater may range from a few hundred to several thousands of years, depending on the type of waste to be stored. Predictions of the long-term corrosion rate and behaviour of iron and steel are based mainly on short-term laboratory experiments and studies of archaeological artefacts. Such predictions are limited by the relatively short timescale (up to c.3000 years) over which these observations are relevant compared to the safety assessment period. Observations from natural analogues may potentially provide information over longer timescales. However, native iron or iron alloys occur only rarely in nature. This paper describes the corrosion of small grains of native iron from a Palaeocene impactite on the Isle of Skye. The deposit was initially buried beneath a thick sequence of Palaeocene basalt before its exposure to subaerial weathering following Devensian glacial erosion. This natural analogue provides an insight into the corrosion of iron, preserved in a sandy clayey matrix, over a timescale extending to c.61 Ma.

Keywords: Natural analogue, radioactive waste, iron, steel, corrosion

Natural Analogues and long-term evolution: upscaling towards repository relevant space and time scales

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Abstract

In the context of radioactive waste management, Natural analogues (NA) can be used to i) upscale data (in space and time) obtained on laboratory and/or underground research laboratories (URL's) in order to test future scenarios of long-term evolution and to build confidence in the safety case. Within the framework of the European Joint Programme of Radioactive Waste Management a workpackage proposal is currently being developed. Promising analogues relevant to near- and far-field processes are under discussion including: i) verified permafrost (PF) models based on polygonal networks in past and present-day PF areas, and depth indicators in the geological record and present-day PF regions. ii) Constraints on landscape evolution scenarios (incl. geodynamic movements, faulting, topographic evolution) at the European scale based on geo(morpho)logical archives and modelling, iii) Constraints on kinetics of rock-water interactions under elevated temperature, through data-model comparison in relevant geological contexts with regard to host rock material and temperature. iv) Constraints on corrosion behaviour (multi-scale) of a) glass including devitrification and b) metals (carbon steel, copper). v) Constraints on flow and transport in host rocks and surrounding aquifers through groundwater dating and tracing of natural study cases in relevant geological settings. vi) Constraints on the performance of soil covers as (hydraulic) barriers for surface disposal through the analysis of natural soil profiles in relevant pedological and hydrogeological settings. vii) Constraints on microbiological and mineralogical processes at the clay-metal interface.

Keywords: landscape evolution; permafrost; corrosion; flow and transport; soils covers

Natural analogues for long-term climato-tectonic evolution scenarios

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Abstract

Typically, the long timeframes (> 100 000 years) that are involved in the safety assessment of radioactive waste disposal systems necessitate the inclusion of external and internal geodynamic processes. Here, we aim at presenting several natural analogues that are relevant with respect to estimating the magnitude and rate of geodynamic processes, and/or estimate the effect of such processes on the geosphere evolution or other components of the disposal system.

The first category includes those analogues that can be used to investigate a natural system under a certain forcing that is expected to take place (or cannot be ruled out) in the future. Typical examples include the study of (i) hydrogeological systems under current permafrost conditions in, e.g., Alaska or Canada, or, (ii) under glacial loading in, e.g., Greenland.

Another category includes those that are useful to determine the rate and/or magnitude of geodynamic processes from the past, and their net effect. Typical examples here are (iii) the determination of past permafrost based on (sub)surface relicts, (iv) the magnitude of glacial deformation of overburden and/or host rock as observed in the geological record (glacio-tectonics, tunnel valleys, glacial basins), (v) the impact of glaciation on host rock properties on formerly glacially-loaded host rock, and, (vi) the rate of uplift/subsidence and erosion based on uplifted and/or buried river terraces.

Finally, (vii) evolved natural soil profiles can be useful analogues for earth covers of surface disposal facilities to determine the effect of soil evolution on, e.g., barrier integrity under the current or different climate.

Keywords: climate evolution; tectonics; erosion; hydrogeology; soil development

Natural glass alteration under a hyperalkaline condition for about 4000 years

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Abstract

Silicate glasses are durable materials in our daily life, but glass corrosion rate accelerates under alkaline aqueous environment. Such situation has raised concerns in nuclear waste disposal where vitrified radioactive wastes encounter to alkaline leachate from surrounding concrete materials. Since glass alteration behavior cannot be ensured under hyperalkaline conditions beyond a few thousand years, it must be examined using geological or archaeological analogues, as along with various laboratory experiments that assess reaction mechanisms at glass-water interfaces. Here we report volcanic glass example surviving with a hyperalkaline groundwater (pH > 11) and high flow rate for about 4000 years. The study area is in Narra, in central Palawan (9˚12'14″N, 118˚16'51″E, ~ 70 m above sea level), where alluvial fan deposits spreading on gentle slopes of serpentinite basement. Volcanic ash layer was found between serpentinite sediments and its depositional age was constrained between 4516±73 and 3445±73 years before present based on 14C dating. The tiny glass fragments were extracted from the volcanic ash layer using a focused ion-beam apparatus, then examined by (scanning) transmission electron microscopy. Sharp elemental distributions were observed at the glass surface, where amorphous-like smectite precursors and crystalline smectites coexist, suggesting the corrosion by an interface-coupled dissolution-precipitation mechanism rather than inter-diffusion which makes passivating layer. The corrosion rate was maintained at, the minimum, 2.5 orders of magnitude less than the rate observed for fresh glass, even in the presence of Fe and Mg that might have consumed Si through silicates precipitation.

Keywords: Nuclear glass, geological disposal, smectite, alkaline alteration

Recent activities on natural analogue studies in Japan

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Abstract

Safety of geological disposal of radioactive waste will be demonstrated in a logical manner by way of a "safety assessment", together with "supporting evidence", such as that provided by natural analogues, that will reinforce/support the assessment results. The extent to which repository safety functions can be validated by short timescale laboratory studies of subsystems or in-situ experiments in URLs is inherently limited. Such experiments are used to the extent possible to test the assumptions, models, and data sets used to assess safety. However, these need to be extended using natural analogue test cases that cover appropriate timescales and the complexity of the natural environment. Overall, natural analogues support the main conclusions of the high isolation potential of a robust system of engineered barriers located in a suitable deep geological setting in Japan. However, these analogues highlight limitations in the models used – in particular in terms of overly simplistic hydrogeology and assumption of thermodynamic equilibrium. While the models tend to be conservative, lack of realism will limit their application to compare different sites and associated repository concepts, as will be needed as the NUMO programme advances. Development of better models will also require appropriate analogues to test them, which is noted as a future goal. In the presentation, the latest analogue studies on the longevity of the carbon steel of the overpack and bentonite buffer will be presented.

Keywords: safety case, natural barriers, engineered barrier system, natural analogue

Microbiology of barrier component analogues of a deep geological repository

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Abstract

Deep geological repositories (DGR) are a solution for long-term nuclear waste storage currently being researched by many countries. As a DGR must remain functional for a million years, studying the microbiology of ancient natural systems that can serve as analogues to DGR design components can provide insight into the microorganisms that could be present and active under DGR conditions. Of particular importance are microorganisms with metabolisms that could contribute to corrosion of metal components of used fuel containers, such as sulfate-reducing bacteria (SRB), or those that produce gases that could potentially produce transport pathways for gases and liquids within a DGR. Ten hand samples from the Tsukinuno bentonite deposit (Japan) were studied using a combination of culturing and DNA-based techniques to explore the abundance, viability, and composition of microbial communities within the clay. Abundances of culturable microorganisms were low in all samples, and sulfate-reducing bacteria were only detected in a single sample. Analysis of 16S rRNA gene amplicons revealed a dominance of sequences associated with putative fermenting bacteria and acidophilic sulfur reducers, though these sequences could represent relic DNA rather than living, viable microorganisms. Future research will involve studying pristine clay cores, with a focus on exploring the viable portion (i.e., without the influence of relic DNA) of the microbial communities using DNA-based techniques.

Keywords

Natural analogue, bentonite, microbiology, culturing, 16S rRNA gene analysis

Michigan International Copper Analogue (MICA) project – Phase I

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Abstract

One of the key requirements for the geological disposal of radioactive waste is assessment of its long-term performance. Observations made from the geological systems can be utilized in the safety case. The Michigan International Copper Analogue (MICA) Project focuses on the stability of copper in the world's largest native copper deposits of the Keweenaw Peninsula, Michigan, USA. The goal is to provide a unique data source to describe natural processes governing long-term corrosion behaviour of the copper used in waste canisters.

The native copper district occurs within the ~1.1 Ga North American Midcontinent Rift system where hydrothermal fluids precipitated native copper and associated minerals in open spaces of the rift-filling host rocks. Subsequent erosion resulted in exposure of the deposits at the surface. The Keweenawan deposits were commercially mined from 1845 to 1968.

In MICA Phase I, geologic setting and environment history was used to group copper occurrences into thirteen analogues for which the history, environmental condition, length of exposure, uncertainties and assumptions were presented based on existing data. The feasibility of representative sampling was assessed for each analogue. Native copper in bedrock with limited exposure to waters provides a baseline level of corrosion. Corrosion analogues identified as the most promising are: 1) bedrock native copper with varied history; 2) cumulative corrosion in native copper of glacial sediments; 3) native copper in fracture-filling clay; and 4) copper exposed to copper sulfides. Phase I provides a starting point to further investigate the corrosion of native copper during Phase II.

Keywords: radioactive waste; final disposal, native copper, Keweenaw Peninsula, USA

PRESENTATIONS

All presentations from the workshop are available on the NAWG website at https://www.natural-analogues.com/nawg-workshops/nawg-17-2023-croatia.