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# Natural Analogues for Safety Cases of Repositories in Rock Salt

Salt Club Workhop Proceedings 5-7 September 2013 Braunschweig, Germany





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#### NUCLEAR ENERGY AGENCY

#### **Radioactive Waste Management Committee**

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"Salt Club" Workshop Proceedings

Braunschweig, Germany 5-7 September 2013

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#### NEA/RWM/R(2013)10

Radioactive Waste Management

## Natural Analogues for Safety Cases of Repositories in Rock Salt "Salt Club" Workshop Proceedings

Braunschweig, Germany 5-7 September 2012

Hosted by

Gesellschaft für Anlagen und Reaktorsicherheit (GRS) mbH Braunschweig

and

Project Management Agency – Water Technology and Waste Management (PTKA-WTE) Karlsruhe

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#### **Foreword**

The Organisation for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA), under its Radioactive Waste Management Committee, allowed its technical working group, the Integration Group for the Safety Case (IGSC) to establish a subsidiary, focused working group on "Safe Disposal of Long-Lived and Heat Generating Radioactive Waste in a Deep Geological Repository in Rock Salt", the so-called Salt Club. The Salt Club was set up by the IGSC in late 2011 and recommended to its supervising organisation the RWMC. The RWMC in turn confirmed the Salt Club as an NEA activity in early 2012.

The overall aim of the Salt Club is to share knowledge relevant to all aspects of radioactive waste disposal in rock salt. Specific areas of work to be addressed include technology and engineering issues, topics addressing geotechnical barriers, and long-term geomechanical and geochemical evolution processes. All of these topics, and more, are components of a long-term post-closure safety case for a repository in rock salt.

Due to the uncertainties in the safety assessment growing with the extended geological time frame, multiple lines of evidence supporting the experimental and modelling results as well as the conclusions to be drawn, will be of increasing importance for the safety case. Information from natural analogues represents one category of supporting arguments. It was recognised that fewer analogue studies have been undertaken in rock salt than in other rock types, such as crystalline rocks or clays.

In view of this one of the first technical activities of the new Salt Club was to plan and conduct an international workshop on natural analogue information related to rock salt as a host rock. This workshop was convened in September 2012 in Braunschweig, Germany. Its participants were charged with identifying and compiling information on known and prospective analogue systems in rock salt. These proceedings allow others to review what was presented, and document the conclusions and recommendations agreed upon at the workshop.

#### Acknowledgements

These proceedings were prepared by Bill Miller (Edinburgh) and Ulrich Noseck (GRS, Gesellschaft für Anlagen- und Reaktorsicherheit, Germany).

Special thanks are also due to:

- the Project Management Agency Karlsruhe Water Technology and Waste Management (PTKA-WTE), the Ministry for Economics and Technology (BMWi) and the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, which hosted the workshop within the frame of the Salt Club activities;
- $\cdot \hspace{0.1in}$  the chairpersons of the various sessions, who led the debates;
- · the speakers for their interesting and stimulating presentations;
- · all participants for their active and constructive contributions.

#### **Table of Contents**

Introduction	9
Scope and Objectives of the Workshop	11
Synthesis of the Workshop	13
Session I: General Aspects of Analogues	27
W.R. Alexander, I.G. McKinley, H. Kawamura  The process of defining an optimal natural analogue programme  to support national disposal programmes	29
C. Pescatore Analogues, analogies, anecdotes and more – Findings of the NEA Forum on Stakeholder Confidence	
Session II: Overview of National Programmes	49
A. Van Luik, R. Nelson  The US government approach to management and geologic disposal  of radioactive waste in the USA – 2013	51
The current status of the geological disposal of radioactive waste in Poland  E. Verhoef, E. Neeft  Status of R&D programme on geological disposal of radioactive waste in	57
the Netherlands	
Session III: Integrity of Rock Salt	85
J. Wolf, U. Noseck, W. Steininger  Applicability of natural analogues for a safety case in salt	
N.T. Rempe  Geologic analogues for hot waste and for radionuclide releases  G. Desbois, S. Li, F. Strozyk, S. Abe, H.van Gent, J. Schmatz, P.A. Kukla, J.L. Urai  Deformation mechanisms and constitutive behaviour during long-term	99
creep of evaporites	105
M. Dulinski, K. Rózánski, K. Brudnik, P. Kolonko, J. Tadych Isotope monitoring of water appearances in salt mines: The Polish experience	123

#### NEA/RWM/R(2013)10

	J.L. Urai, P.A. Kukla, G. Desbois, J. Schmatz, M. Sadler, Z. Schléder, J. Schönherr	
	Fluids and fluid migration in salt	133
	J. Hammer, M. Mertineit, J. Behlau, G. Mingerzahn,	
	T. Kühnlenz, M. Schramm, G. Zulauf, J. Zulauf	
	Deformation and geometry of anhydrite rocks in domal salt	
	structures – results of field studies, mineralogical analyses and	
	thermomechanical experiments	147
	W. Minkley, M. Knauth	
	Integrity of rock salt formations under static and dynamic impact	177
	J.L. Urai, P.A. Kukla, F. Strozyk, H. van Gent, S. Abe, S. Li, G. Desbois,	
	J. Schönherr, Z. Schléder, L. Reuning, S. Becker	
	Internal dynamics of salt structures	197
Se	ssion IV: Long-Term Properties of Geotechnical Barriers	205
	U. Noseck, J. Wolf, F. Crotogino	
	Geotechnical barriers in the repository concept for HLW disposal in	
	rock salt	207
Se	ssion V: Analogues for Chemical and Microbial Processes	243
	V. Metz, Y.O. Rosenberg, M. Böttle, J. Ganor	
	Ra-Ba precipitation in laboratory and large-scale evaporitic systems	245
	G. Bracke	
	Consideration of hydrocarbons at the Gorleben salt dome in a preliminary	
	safety assessment	253
	A. Meleshyn	
	Analogues for microbial effects in rock salt	259
	ringly such in initial chieff in i for suit	

#### Introduction

Rock salt has long been considered as a potential host rock for geological disposal of radioactive wastes. The WIPP repository (New Mexico, United States) is constructed in rock salt and is actively being used for the disposal of long-lived transuranic waste. The Morsleben repository (Saxony-Anhalt, Germany) constructed in rock salt was used for the disposal of low- and intermediate-level wastes between 1971 and 1998, and is now being decommissioned. Several other disposal programmes are also considering rock salt as a potential host rock for geological disposal of high-level and long-lived wastes because of its favourable characteristics.

In 2012, the OECD/NEA established a new IGSC working group on "Safe Disposal of Long-Lived and Heat Generating Radioactive Waste in a Deep Geological Repository in Rock Salt", the so-called Salt Club. The overall aims of the working group are to improve understanding and to share knowledge relevant to all aspects of radioactive waste disposal in salt. Specific areas of work to be addressed include technology and engineering issues, topics addressing the geotechnical barriers, long-term geomechanical and geochemical evolution processes, and long-term post-closure safety case issues.

It is not possible to simulate in laboratory studies the very long-term process that might affect the safety performance of a repository. For this reason, natural, archaeological and industrial analogue studies are often used as one of several multiple lines of reasoning that, when combined, help to build understanding and confidence.

Salt has been mined as an economically important resource for hundreds of years and, in the last several decades, has been intensively studied because of its geological association with oil and gas. As a consequence, there are many sites and environments world wide that offer potential as analogues relevant to some aspects of geological disposal systems in rock salt. For this reason, it was decided that one of the first topics addressed in the working agenda of the Salt Club should be natural analogues.

As an early activity of the Salt Club, an international workshop was convened in September 2012 in Braunschweig, Germany, with the purpose of identifying and compiling information on known and prospective analogue systems in rock salt. These proceedings provides a record of that workshop and of the conclusions and recommendations that were reached.

#### NEA/RWM/R(2013)10

#### Scope and Objectives of the Workshop

The workshop on "Natural Analogues for Safety Cases of Repositories in Rock Salt" had the overall objective of compiling studies about natural as well as anthropogenic analogues from different countries to be potentially used within safety cases for radioactive waste repositories in salt formations. This objective enabled the broad variety, number and nature of potential analogues in salt to be established.

The scope of the Salt Club covers long-lived and heat-generating waste disposal in salt. For this reason, the workshop paid specific attention to long-term processes in salt and to processes that may be affected by high temperatures or thermal gradients. Presentations and discussions took place around the topical areas of:

- long-term integrity of rock salt formations;
- · integrity of technical barriers;
- · microbial, chemical and transport processes.

An important objective of the workshop was to promote debate and discussion about the status and potential usefulness of known and potential natural analogue studies in rock salt. For this reason, time was given over in the agenda to working sessions in which participants were encouraged to consider the key unresolved issues and to identify any high priority recommendations for future natural analogue studies in salt, as well as any possible recommendations to the Salt Club.

Workshop participants were drawn from disposal agencies, research organisations, industry and regulatory authorities across Europe and from the United States. Importantly, the participants included researchers (geologists, material scientists, microbiologists, etc.) as well as safety assessors. This interdisciplinary mix of participants stimulated the discussion on the nature of the geoscientific information that can be obtained in field measurements in salt, as well as the data requirements of safety assessment models. Lessons learned from previous projects show that this dialogue is essential for maximum benefit from analogue information to be gained.

Presentations and discussions about research are an essential part of the scientific process, but conclusions need to be drawn to focus future work. For this reason, time was set aside at the end of the workshop to debate the key observations and preliminary findings from the analogue studies presented. These findings are provided in the following "Synthesis of the Workshop" which summarises the analogue studies presented, and also sets out a series of conclusions regarding their collective outcomes. The synthesis ends with a set of recommendations for further work which is focused on certain key unresolved issues that the workshop participants considered to be potentially amenable to further investigation by natural analogues in rock salt.

#### NEA/RWM/R(2013)10

#### Synthesis of the Workshop

#### Bill Miller, Ulrich Noseck, Jens Wolf

#### Introduction

Rock salt is a potential host rock for a geological repository for the disposal of high-level and long-lived wastes because of its favourable characteristics. Due to its very low hydraulic conductivity and its ability to deform plastically under stress, rock salt can, under the right circumstances, provide robust, long-term isolation of the waste from groundwater.

In 2012, the OECD/NEA established under the aegis of the RWMC a new IGSC working group on "Safe Disposal of Long-Lived and Heat Generating Radioactive Waste in a Deep Geological Repository in Rock Salt", the so-called Salt Club, considering all aspects of radioactive waste (HLW and long-lived) disposal in rock salt. It consists of representatives from the founding member states, namely Germany, Poland, the Netherlands and the United States.

The Salt Club has the general objective of effectively developing and exchanging scientific information and shared approaches as well as methods to develop and document an understanding of rock salt as a host rock formation for a geological repository. Mission and objectives are written down in the Terms of Reference (www.oecd-nea.org/rwm/saltclub/). Areas of work comprise e.g. technology/engineering issues, topics addressing the geotechnical barriers, geomechanical and geochemical issues and of course, safety case issues. Natural analogues are explicitly addressed within the geochemical and safety case topics.

It was decided that one of the first topics addressed in the working agenda of the Salt Club should be natural analogues.

Best practice in repository safety cases is based on the use of multiple lines of reasoning (NEA, 2012), and one important line of reasoning for improving repository system understanding is the use of natural analogues.

From 4-6 September 2012 the Project Management Agency Karlsruhe – Water Technology and Waste Management (PTKA-WTE) and the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH hosted within the frame of the Salt Club activities a joint workshop on "Natural Analogues for Safety Cases of Repositories in Rock Salt". The workshop was the first of a series of activities to be addressed by the Salt Club and took place in Braunschweig, Germany.

The objective of the workshop was to compile studies about natural as well as anthropogenic analogues (NA) from different countries to be potentially used within safety cases for radioactive waste repositories in salt formations.

The first part of the workshop programme consisted of four sessions with presentations on:

- · national programmes of the Salt Club member states (the United States, Germany, Poland and the Netherlands) and international NA activities (NAWG, NEA-FSC);
- · the long-term integrity of rock salt formations;
- the integrity of technical barriers;
- · microbial, chemical and transport processes.

In the second part of the workshop four parallel working sessions discussed the status of NA in salt, the key unresolved issues, high priority recommendations for future NA studies for salt and possible recommendations to the Salt Club. The results of the discussion are summarised below. The workshop was concluded with a one-day excursion to the German rock salt repository for LLW/ILW in Morsleben (ERAM).

The workshop attracted both organisations working actively in the field of NA, and also other organisations, e.g. regulatory bodies that wished to be informed on the status and practical applicability of NA, as well as companies from the salt mining and the oil/gas storage industry that presented their practical experiences.

The Salt Club includes natural analogues within its areas of interest, and the findings of the workshop described in this report are a contribution to the Salt Club's work on this topic.

This summary provides observations and some recommendations drawn from the presentations and discussions at the workshop. For the sake of convenience, the term "natural analogue" is used here in its widest sense, and includes anthropogenic analogues where appropriate.

#### General aspects of natural analogues for the safety case

There has been considerable development in thinking with regard to natural analogues over the last decades. Early analogue studies were very often simply geochemical research projects intended to provide field data concerning the processes (and their rates, where possible) that control radionuclide migration, e.g. in and around uranium ore bodies. These studies were often planned and interpreted with limited involvement from experts in safety cases.

More recently, there has been a recognition that natural analogues can positively contribute to multiple lines of reasoning in a safety case, along with other forms of study such as palaeohydrogeology and the use of complementary safety indicators (NEA, 2012). It is now widely accepted that there are multiple ways in which natural analogues can support a modern safety case for geological disposal (Miller, et al., 2006). For example, as part of the assessment basis they can be useful for:

- features, events and processes (FEP) identification and screening, and scenario development;
- conceptual model development;
- data provision;
- uncertainty management;
- · numerical model, code and data testing, and confidence building;
- in a broader sense, confidence building of experts and laypersons.

More generally, natural analogues may be useful for helping compile the evidence, analyses and arguments and to place the safety case and its results into context. Lastly,

there is the possibility that analogues may be useful during communication and dialogue with stakeholders, provided the analogues are meaningful to the audience and appropriately used.

It should be noted that there is a distinction between the identification of a potential study or site, and it being accepted as a relevant analogue to support a safety case. Potential analogues (and analogue-derived information) need to be critically evaluated to ensure their appropriateness to both the disposal concept and safety strategy, as well as to the safety assessment methodology.

Sometimes in the early days of natural analogue studies, interesting geological and geochemical systems were first identified and then attempts were made to compare them (backfit them) to the repository system and the safety case. There was sometimes a lack of rigour when choosing analogues and when applying "quality assurance" checks to the information derived from analogue studies. This led to some cases of misrepresentation of the equivalence between natural and repository systems, and over interpretation of analogue information. In a reversal of this approach, Alexander, McKinley and Kawamura suggest in future a "top-down" method should be followed to identifying natural analogues. In this, goals for natural analogues are first set (such as the goal to demonstrate stability of the host rock) and then appropriate analogue sites are sought or relevant analogue information extracted from the published literature. This goal-setting approach for analogues has some parallels to the recent development of defining safety functions for the disposal system and its components. Setting goals for natural analogue studies may be one way (alongside laboratory and modelling studies) to seek information to help confirm that a safety function is met.

As previously mentioned, many different natural systems and sites, from a number of countries, were discussed at the workshop. Several of these have not previously been recognised or examined as potential analogues. The broad range of examples presented illustrates that the scope of analogues for rock salt is potentially extensive. Without diluting the overarching concept of natural analogues, it may sometimes be useful to recognise the different "types" of analogues, for example:

- Industrial analogues such as gas storage caverns in rock salt that may provide information on aspects to do with the geomechanical behaviour of the rock during excavation of the repository and of technical barriers.
- · Contemporary analogues that may provide information on short-term processes, such as the response of the rock mass during re-establishment of THMC equilibria.
- Operational analogues that provide information on practical aspects of constructing and closing excavations in salt, which are likely to be increasingly useful as programmes move towards implementation.
- National analogues that place the repository into a national or regional context by providing local examples that may be most meaningful to the public and other stakeholders.
- Social analogues that may refer to aspects of demographics and the behaviours, e.g. in decision making, of the current generation that may be analogous to future populations.
- Negative or anti analogues which highlight that processes and rates can be more aggressive and faster in certain environments than is often assumed for repository conditions, i.e. most archaeological evidence suggests iron artefacts corrode away, and are not preserved.
- Self analogues which refers to information derived from the actual repository site, rather than another location.

An important aspect that was stressed from the discussion at the workshop of the possible "types" of analogue studies is the importance of correct and unambiguous terminology to avoid possible misunderstandings between different audiences. This is an issue for the safety case as a whole because it should be recognised that the meaning of certain words used by the public in common speech is sometimes different than the usage by experts (the word "risk" for example), and even occasionally between different groups of experts.

In particular, there can be differences of opinion as to what represents an appropriate analogue site or system, and what does not. The question of "equivalence" with a repository system is fundamental to the concept of natural analogues. It is clear that if analogue information is used in safety assessment modelling, it should be of the highest quality and also that the similarities and differences between the analogue and repository systems should be understood and transparently described.

Decision making throughout a repository development programme is iterative, and different but appropriate levels of information are required at each key stage and decision point. It follows that qualitative analogue information and semi-qualitative analogue data may be used in the early stages of a programme to help underpin certain preliminary decisions. For example, the weight of archaeological evidence on metal corrosion may be used (when combined with laboratory data) to narrow down the choice of potential container materials. At later stages in the repository development programme, however, such qualitative analogue information would be insufficient to support detailed optioneering or design optimisation work, and more quantitative information would be needed.

As suggested by Pescatore, one approach to reflect these different applications of analogues within a repository development programme may be to adopt a hierarchy of concepts that could be represented by using alternative terms (with slightly different definitions) such as "anecdotes", "analogies" and "analogues" to indicate increasing levels of similarity between the natural and repository systems. This is a novel suggestion and one that might be worthy of further consideration. It is worth noting that this suggestion is born partly out of work by the OECD/NEA Forum for Stakeholder Confidence (FSC) that highlighted the importance of terminology for confidence building.

There has long been a debate regarding the potential value of using analogues for public communication and confidence building. The visual appeal and everyday context of some analogues (e.g. the longevity of Roman buildings still standing in many European cities) have often been cited as reasons why analogues could be useful when engaging in dialogue with non-technical audiences. Several national programmes have used analogues in this way in the early days of analogue studies (e.g. Spain, Sweden and the United Kingdom) but, despite this, there is no solid evidence to suggest that they have proved useful for explaining disposal concepts and long-term safety. Part of the nervousness for using analogues for public communication may be related to the concern of "over interpretation" but provided that analogues are used appropriately there seems to be no reason why qualitative analogues should not be used for general confidence building as a parallel activity to using quantitative analogues for technical input to safety assessment.

Given the points raised during discussion at the workshop, it is evident that there are a number of issues that could usefully be addressed in the future. These include:

- · Care should be taken to identify the various roles that natural analogues could play within the overall safety case, understanding that different applications are possible and that each may have their own specific requirements.
- Within the context of safety assessment, analogue studies and analogue information should be critically assessed, with particular attention paid to understanding the implications of the differences between the analogue and repository systems (such as the boundary conditions), as well as the similarities. Natural analogues should

not be considered in isolation. Their value is enhanced when used in combination with other multiple lines of reasoning, such as palaeohydrogeological investigations, when the advantages of one can be used to balance the drawbacks of another.

- Care should always be taken to avoid over interpretation and abuse of analogue information. In the context of preliminary decision making and for concept development, the requirements for analogues may be more relaxed than for detailed safety assessment modelling.
- The use of analogues for public communication and dialogue remains unproven, and is an area worthy of further consideration. It is likely that analogues with a familiar and local context may be most useful in this regard.

### The application of analogues in national repository development programmes based on rock salt

The various national repository development programmes around the world investigating disposal in rock salt are advancing at different rates. This is due, in part, to variations in government policy but also the practicalities of identifying suitable geological environments, host communities and progressing the underpinning technical work. Advanced programmes are the WIPP site (New Mexico, United States) and Morsleben (Saxony-Anhalt, Germany). These disposal sites vary, however, insofar as WIPP remains operational but Morsleben is to be decommissioned. A number of other national programmes have identified rock salt as a preferred host rock but have not yet chosen a candidate site (e.g. Poland), and others retain rock salt alongside crystalline and argillaceous rocks as a possible host rock type in an open siting process (e.g. the United Kingdom, the Netherlands, Belarus).

The use of natural analogues in disposal programmes based on rock salt to date is also variable. As one example natural analogues were undertaken in support of WIPP to aid system understanding but did not feature directly in the formal safety case for the facility (NEA, 2000). The types of analogue studies that were performed include (Murphy and Kovach, 1993):

- · uranium migration studies in the fractured Culebra dolomite aquifer that overlies the bedded salt deposits that host the repository;
- earthquake activity and seismic impacts (rock bursts in salt mines);
- material corrosion rate assessments based on preservation of artefacts found in old salt mines.

Although natural analogues did feature within the WIPP project, in general, fewer analogues studies have been undertaken in rock salt (Noseck, et al., 2008; Wolf, et al., 2013) than in other rock types, such as in crystalline rocks (Miller, et al., 2006; Laverov, et al., 2008). The reason for this is not entirely obvious, but probably simply relates to the fact that the national programmes that initially developed the concept of analogues in the 1980s (at least in Europe, notably Switzerland, Sweden and the United Kingdom) were not focused on rock salt.

Germany started investigations into NA in the mid-nineties after a national workshop considering this topic was organised. This workshop discussed some R&D projects that had been conducted, as well as some natural analogues that had already been running for several years such as the Ruprechtov NA in the Czech Republic (Noseck, et al., 2008a, 2009). After the workshop, a comprehensive study on the use of NA was undertaken (Noseck, et al., 2008b; Wolf, et al., 2013) and presented.

Important outcomes of that workshop were the conclusion to proceed systematically in the use of NA, and the identification of many different natural systems and sites, from different countries, which expands the portfolio of analogue studies that are potentially relevant to safety cases for disposal in rock salt.

Looking to the future, there is likely to be greater interest and use of analogues within disposal programmes based on rock salt following the Salt Club initiative. As an example, Wolf, Noseck and Steininger explain how Germany is preparing for a new strategy that intends to use natural analogues as an integral component in its safety case methodology. As an early part of the process, a comprehensive compilation and assessment of natural analogues relevant to rock salt is under way. An interesting and unique aspect of this work is an attempt to categorise analogue studies in a structured manner according to their relevance to the important elements of the German safety and assessment concept: i) the integrity of the geological barrier; ii) the integrity of the geotechnical barriers; iii) radionuclide release scenarios. The availability and quality of study reports is also assessed to identify which analogues are sufficiently well documented to be of possible value for either technical confidence building or public confidence building.

The WIPP and Morsleben repositories provide an opportunity for learning based on experience from construction and operation of these facilities. This is a form of "implementation analogue" albeit more applicable to practical operations than development of the safety case. Various lessons learned from the WIPP operating experience that could be applicable to other repositories include such things as to avoid mixing different wastes in the same vault, and to adopt a flexible programme of cavern excavation and waste emplacement, to mitigate against delays in waste shipments, etc.

Finally, a particular feature of rock salt is that formations can, in some cases, be laterally extensive and internally homogenous. In Europe, certain formations extend across national boundaries, such as the Zechstein which extends from across northern Europe from the United Kingdom to Poland. This provides an opportunity for increased international co-operation and research into analogue studies related to disposal in rock salt.

#### Rock salt specific analogue results

The primary characteristics (and safety functions) of rock salt that contribute to long-term isolation of the waste from the accessible environment are: i) very low hydraulic conductivity; ii) the ability to deform plastically under stress, so discontinuities, which might present potential pathways, can "self-seal". These are somewhat different to the characteristics of other potential host rocks that provide isolation by processes such as radionuclide sorption, dilution and dispersion along groundwater flow paths. As a consequence, natural analogue studies relevant to rock salt are very often focused on containment processes rather than transport/retardation processes.

Most analogues studies performed so far on rock salt have investigated processes that may impact on the integrity of the salt and cause a reduction in its containment performance. These studies seek to improve conceptual understanding of relevant THMC process mechanisms, couplings and boundary conditions, as well as to quantify their rates and limits. A number of studies were presented and discussed at the workshop, and some highlights from these are given below:

Thermal processes have been investigated around volcanic dyke and sill intrusions into rock salt. These may be analogous to the heat generated by vitrified HLW and spent fuels, although the temperatures in a repository near-field (a likely maximum of c. 100-200°C depending on the repository design) will be much lower than experienced in a contact metamorphic situation (c. 1 000°C). As explained by Rempe,

natural analogues thus provide practical examples of worst case scenarios in which the containment performance of rock salt would not be compromised.

Hydraulic processes have been investigated in several analogue systems. Observations and measurements of water inflows in Polish salt mines presented by Dulinski illustrate the potential use of isotopes to characterise the age and origin of brines in salt formations. On that basis syngenetic (intrasalinar) water appearances can be clearly distinguished from recent (Quaternary) and modern waters. Such measurements are important to identify the source of waters not only in salt mines, which have been excavated for several tens to hundreds of years, but also in salt repositories where water intrusion from the adjacent geologic formations can impact the safe operation of the mine and have to be investigated. Urai illustrates that free water can be present in rock salt on spatial scales ranging from micrometre sized fluid inclusions to tens of metres sized brine pockets. A key observation of this work is that fluid inclusions may be separate and immobile under normal conditions but can become mobile if the stress conditions change. Accumulations of brine in larger pockets can also rise upwards through salt deposits, driven by buoyancy. Similarly, oil from big intrasalinar carbonatic rock inclusions has been observed to migrate through deformed and intensively uprising salt over large distances, as observed in oil deposits in Oman. This salt dome is characterised by a high content of hydrocarbons and is in the very active diapiric phase. However, this behaviour cannot be observed in the salt domes of the Northern German Zechstein Basin in the post-diapiric phase. The brine pockets around the WIPP facility are also observed to be immobile due to the static geological conditions.

- Mechanical processes in rock salt have been investigated at numerous sites and on different spatial scales. On a large scale, 3-D seismic modelling of salt domes by the oil and gas industry, and described by Urai, show the influence of the anhydrite layers that are mechanically stronger than the associated halite in the salt dome structure. This is supported by laboratory studies, such as those described by Mertineit, Hammer, Zulauf, Zulauf and Schulze, that show when subject to stress, halite will deform by plastic flow but anhydrite will deform by brittle fracture. Because of the creeping of rock salt the fractures healed, and consequently the permeability of the rock remains very low (Hammer, et al., 2012). In a repository environment, the safety case must show that these fractures are not possible pathways for groundwater flow. Detailed observations from exposures and boreholes at the Gorleben salt dome and Morsleben repository presented by Hammer, Behlau, Mingerzahn and Mertineit showed, however, that the anhydrite at Gorleben and Morsleben (Hauptanhydrite, z3HA, Gorleben-Bank, z3OSM and Anhydritmittelsalz, z3AM) do not contain large-scale interconnected porosity, but clear evidence of secondary sealing in the fractures. This is a good example of how laboratory, field and analogue studies can be combined to increase process understanding and, in particular, the issue of spatial scaling.
- Microbial populations have been recorded in rock salt, including in mines and in brine inclusions, as described by Meleshyn. It is probable, therefore, that microbes will be present in the near-field of a repository but it is unclear if they will be viable in the post-closure environment and affect repository performance (e.g. by the production of gas and corrosive substances) and, consequently, whether they need to be explicitly considered in the safety case. Laboratory and field studies (e.g. at WIPP) confirm the presence of ancient indigenous microbes as well as modern introduced species but, to date, few analogue studies have provided convincing data on their viability under repository post-closure conditions.
- · Certain chemical processes that may affect the safety performance of a repository might potentially be more significant in rock salt than in other geological environments. Of particular interest are the possible consequences of reactions

involving hydrocarbons because of the common geological association between oil and gas deposits, and salt domes. Hydrocarbons can be dispersed throughout salt and readily detected in mines, such as at Gorleben, as illustrated by Bracke. During repository operations, accumulation of flammable gas may increase the risk of explosion. It must be dealt with in the construction and operational safety assessments for the disposal facility. In the post-closure period, thermochemical reduction of sulphates by hydrocarbons has been postulated as a mechanism that may lead to production of water and corrosive substances that could lead to a loss of integrity of the rock salt. As with microbial processes, however, few analogue studies have provided information on the likelihood and consequences of these processes occurring under repository conditions.

Releases of <sup>226</sup>Ra often dominate the calculated post-closure doses in repository safety assessments due to the assumption of high mobility and the corresponding choice of conservative values for solubility and retardation factors in transport models. Nonetheless, as explained by Metz, Rosenberg, Böttle and Ganor, there is analogue evidence to show that Ra can often be retarded by co-precipitation with Ba within (Ba,Ra)SO4 (barite) solid solutions in crystalline and argillaceous environments. More recent studies now indicate that Ra can also be retarded by co-precipitation in saline systems, such as the Ketziot desalination plant in Israel. These findings are generally supported by appropriate laboratory experiments. However, differences in partition coefficients measured in co-precipitation and re-precipitation experiments, respectively, need to be resolved in further studies, to determine the extent to which the radiobarite co- and re-precipitation systems approach near-equilibrium conditions. Other studies would be useful to extend these observations to systems that more closely resemble the repository environment to determine whether co-precipitation could be taken into account in the safety case for a repository in rock salt to reduce the conservatism in the transport models.

It is also clear that the coupling of THMC processes in rock salt is complex, with multiple potential mechanisms that could affect the integrity of rock salt as a geological barrier. Nonetheless, there is also strong evidence from natural systems to confirm that rock salt is a stable environment, and so may provide long-term containment for radioactive wastes, provided that certain boundary conditions are not exceeded. It is clearly important to define the thresholds between a stable and a dynamic situation, and to understand what FEPs may cause a change to occur. These may potentially be internal to the repository system (e.g. a build-up in gas pressure due to corrosion or microbial action) or external (e.g. a change to groundwater flow rates and chemistry driven by climate evolution).

Many processes occurring in rock salt do so over a range of spatial and temporal scales. Notably, fluid migration can occur on the microscopic (grain boundary) to macroscopic (brine pocket) scales. It is important to understand how analogue and laboratory observations could be scaled to the repository environment and assessment time periods, recognising that scaling relationships are unlikely to be linear.

Natural analogue studies alone cannot answer these questions, but they may provide strong collaborative evidence, together with laboratory and modelling studies. When seeking new analogue studies and sites, and when evaluating analogue information, those that have specific relevance to the safety functions and characteristics of rock salt would be most appropriate to consider, recognising that these are different to other rock types, and so may require unique analogues. In particular, analogues that provide information on the low hydraulic conductivity and self-sealing properties of rock salt would be particularly appropriate. In addition, analogues that shine light on the dynamic processes relevant to evolution scenarios for the repository near-field are particularly valuable, most notably those that might provide information on scenarios that could trigger groundwater movement over a large scale.

The studies presented and discussed during the workshop have proven particularly useful for improving the qualitative understanding of processes that occur in rock salt. This is valuable for the safety case and helps build confidence in the conceptual models that underpin the numerical models. The greater challenge for analogue studies is to provide quantitative information that might help set conservatively realistic limits to specific parameter values used in the numerical models, or to help define the thresholds (limits) to environmental conditions that if exceeded cause a change to more dynamic behaviour.

With this in mind, analogues for rock salt should be drawn from a wide range of relevant situations, including modern industrial examples as well as from geological systems, as discussed below.

#### **Learning lessons from other industries**

Observations from the analogue studies described above give confidence that rock salt can provide long-term containment for radioactive wastes, provided that certain boundary conditions such as temperature, stress, strain and pressure are not exceeded. In addition to the geological barrier, geotechnical barriers like plugs and seals play an important role in the safety concept for repositories in rock salt. The sealing design for the German safety concept is based on backfilling of the open void volumes in the emplacement areas after disposal of the waste with crushed salt. The cross drifts of the disposal areas will be sealed with seals against the main drifts and the transport drifts with seals against the infrastructure area. Shaft seals are needed to prevent water inflow through the shafts. Since credit for these seals is taken for several thousand years in the safety case, analogue information could provide strong arguments for the integrity of such barriers over longer time scales than addressed in laboratory experiments.

An understanding of the nature of the boundary conditions and thresholds mentioned above comes from experience from other large-scale excavations in salt, such as mines and gas storage caverns, as described by Knauth and Minkley and also by Crotogino. These industrial analogues confirm that rock salt is impermeable to groundwater and gas flow, provided the salt remains undisturbed. There are, however, numerous examples where changes to boundary conditions have caused a loss of integrity, leading to collapse of excavations, damage to surface structures and groundwater to flow into tunnels, etc. Modelling studies show that this situation may arise if either the minimum stress or dilatancy criteria are exceeded.

Experiences from disposing of hazardous wastes in salt and potash mines were described by Lukas. These industrial analogues provide very useful practical information on aspects such as the design of tunnel backfills, plugs and seals. There are now a number of hazardous waste disposal cells, and associated access shafts, that have been closed and sealed (using bentonite and gravel seals keyed into the salt), and these may provide unique opportunities for performance confirmation monitoring of the seals over several decades into the future, and to help in the design of the seals and plugs for a radioactive waste repository. Although clearly very informative, the transferability of industrial analogue information to the repository system needs to be carefully assessed. As is the case with all forms of analogues, the similarities between the conditions of industrial analogues and the repository conditions need to be identified and evaluated, as well as the differences.

For example, there may be different views regarding the stability of excavations in rock salt, and its integrity as a barrier, due to the very different time scales over which it needs to be assessed for disposal compared to other situations. As a consequence, it cannot be assumed that practices (e.g. modelling approaches and facility designs) adopted in one industrial situation can be directly applied to the repository. They can, however, provide a very useful starting point, and one that may avoid duplicating effort. This means that

where there is overlap and areas of common interest between radioactive waste disposal and other industries, there may be clear benefits to be gained from cross-industry liaison, collaborative research and exchange of knowledge and experience. This will help to improve conceptual and numerical models for rock salt behaviour, and assess its integrity as a barrier over the long term.

#### Final comments and conclusions

This workshop within the scope of the NEA's Salt Club represents a rare attempt to identify and evaluate natural analogues specific to disposal concepts in rock salt. The significant differences between the characteristics and safety functions of rock salt compared to other potential host rocks means that there is considerable benefit to this focus. In particular, it is important that analogues for rock salt are found that are relevant to containment processes rather than just transport processes, which has usually been the case for analogues for crystalline and argillaceous host rocks.

Rock salt can be a dynamic environment, with coupled THMC processes occurring that may affect the long-term safety performance of a repository. Natural analogues can, therefore, be equally important for aiding FEP screening and scenario development in the early stages of a safety case, as they are for supporting the subsequent safety assessment modelling calculations.

It is important to realise, however, that there is a difference between observing a process in an analogue system and concluding that the same process will occur and be significant in the repository system. There are many cases where processes seen in nature may be only of secondary importance to the safety of the repository. There are yet others where the potential significance of processes remains an open question. Examples discussed at the workshop include microbial populations and hydrocarbon accumulations; the importance of both for a repository in rock salt is open to debate.

For this reason, analogues need to be fully integrated with the safety assessment modelling strategy. It will often be the case that a natural analogue study will demonstrate that a process may occur under repository conditions but that a mechanistic model may be required to determine its significance.

In the early stages, when a programme may be seeking to identify disposal concepts and reference designs and materials, qualitative information may be sufficient. As the programme proceeds towards design optimisation and formal safety assessments, then there will be an increasing need for more quantitative information.

This raises the concept of "fit for purpose" analogue information. Whilst quality assured work should always be an aim, this is different to treating every analogue study as an academic research project. The objectives of an analogue study should be clearly defined and related to information needs. These needs may vary depending on the status of the repository development programme, and reflect that decision making throughout the programme will be iterative and become progressively more detailed over time.

It follows that there is a challenge to all natural analogue studies to progress beyond simple qualitative observation to aid conceptual understanding, and aim to deliver more quantitative results that might help set the parameter values used in the numerical models, or to define the boundary conditions for system behaviour.

For this to be achieved, it will be essential that:

- · Future analogues be based on high-quality science.
- · The similarities and differences between an analogue system and the repository system be critically assessed.

- These similarities and differences be clearly explained when the analogue study is described and interpreted.
- All analogue information used to support the safety case should be properly justified.

Natural analogues for rock salt should not, however, be considered in isolation but are best treated in the safety case in combination with other multiple lines of reasoning, such as palaeohydrogeology and complementary safety indicators. The reason for this is that no single line of argument can be conclusive, but by combining different arguments, the benefits of one may offset the disadvantages of another.

When seeking new analogue studies and sites, it is particularly useful to have a broad view, and to also seek analogues from modern industrial systems (such as gas storage, mines and oil/gas exploration) as well as from the more traditional natural geological systems. In particular, industrial analogues may provide information e.g. on the response of rock salt to hydromechanical perturbations, and help to define the mechanisms that can cause rock salt to stop behaving as a static material and begin to act in a dynamic manner, and may be used also to quantify the threshold at which that change happens. Industrial analogues (e.g. experience from ongoing hazardous waste disposal operations) may also be useful for addressing a number of practical issues such as backfilling, closure and sealing of a repository, and methods for waste retrieval should that be necessary.

There is clearly benefit to be gained from exchange of knowledge between scientists/ experts and industries involved in rock salt. A number of issues may be of common interest, such as the need to model the behaviour of rock salt and assess its integrity as a barrier.

Despite several decades of analogue study, the value of natural analogues for public communication and dialogue remains unproven. Given that analogues are widely accepted as a significant contributor to the safety case, and that a primary role of the safety case is to build confidence, it is somewhat surprising that no national or international agency has yet undertaken any serious attempt to test analogues as a communication tool with non-technical audiences. Intuition suggests that appropriately chosen analogues that provide meaningful context (e.g. regarding site, materials and time scales) would be useful to help present the safety case to a range of stakeholders.

#### **Recommendations for further work**

In this section, some recommendations for further activities regarding natural analogue studies relevant to rock salt are provided, on the basis of the discussions and comments made at the workshop. This is not intended to be a comprehensive list of all necessary future work, but rather to provide some signposts to activities that may bring early benefit to workers in the field.

It is first recommended that all organisations with an interest in analogue studies undertake a systematic review of their key information needs (a gap analysis), and then decide which of these gaps may best be fulfilled by analogue information. In this context, the "needs" are likely to be varied and depend on the stage of the repository development programme and so the required "information" may take different forms, including both hard, numerical data and soft, qualitative observation. The point is clearly to determine the objectives and success criteria for future analogue studies so that they can be better planned.

At an international level, a good starting point for this gap analysis is the Salt Club's planned FEP Report. It is recommended that this report go beyond simply describing FEPs but also make an attempt to identify where analogue information (and other sources of data) exist to support both conceptual understanding and numerical parameterisation of each individual FEP. If no analogue information can be identified, or the existing information is inadequate, then this may suggest an opportunity for a new study. However, it is not credible to suggest that an analogue is needed or available for every FEP.

The early engagement with other industries demonstrated at the workshop is commended, and steps should be made to collaborate or at least exchange information on future studies when there is a common interest. A first priority could be to identify and understand why there are differences in how the integrity of rock salt is perceived and assessed by different industries, and also by their regulatory bodies (e.g. between those regulating nuclear safety and mining operations). Engagement with other industries will also help to identify sources of existing analogue information and potential future analogue studies.

When planning new analogue studies, or when evaluating existing analogue information, there needs to be full integration with safety assessors. This is an old message but one worth repeating. It is important that analogue information be fit for purpose when applied in the safety case to ensure best value can be obtained from the studies.

Similarly, natural analogue studies should be integrated with other parallel geoscientific and observational studies (such as palaeohydrogeology, natural safety indicators, site characterisation, etc.). There should be synergy between the multiple lines of reasoning used in the safety case so that they mutually support each other, rather than being viewed as separate and unrelated activities. The international agencies may take a lead in this area, by ensuring effective dialogue and information sharing between the ongoing initiatives such as the NEA's Salt Club.

Multiple lines of reasoning are intended to help build confidence amongst all stakeholders, including non-technical audiences. In this context, it is recommended that a rigorous attempt be made to evaluate the true potential of analogues for public communication and dialogue through a structured opinion survey. This is an oft-debated issue, and there are opposing views as to whether or not analogues help build confidence with non-technical audiences or whether they are a distraction. The work by the NEA's FSC in 2008 began to examine this issue but involved only disposal agencies, rather than the wider stakeholder community. It would be useful to the safety case community as a whole to have some guidance on this issue. The NEA may be best placed to undertake this work.

In addition to the general recommendations above, it was evident from discussions at the workshop that there are some specific technical issues that may benefit from further analogue study, and some of these are listed below. The individual papers provided more details on these and other issues. This is not intended to be a "shopping list" for funding agencies but a starting point to identify some significant issues and processes in rock salt where analogue information might reasonably be expected to provide further insight, understanding and quantification:

- Compaction of crushed salt backfill and stability of plugs and seals: The backfill and closure components in a repository need to withstand the slow plastic flow and deformation within rock salt. There are several hundreds of years of experience from salt mine operations and closure that might potentially be useful to the design and assessment of the closure components. Recent sealing of shafts at hazardous waste disposal sites in rock salt provide opportunities for performance confirmation monitoring of a closed facility.
- Deformation and blocking of anhydrite: Detailed observations from exposures and boreholes at the Gorleben salt dome and Morsleben repository showed that following deformation the blocks of former anhydrite layers contain no large-scale interconnected porosity, but clear evidence of healing the fractures. Laboratory, field and analogue studies can be combined to increase process understanding and, in particular, the issue of spatial scaling. Examples from other salt domes would provide strong arguments for a safety case underpinning that this process is likely to occur.

- Isotope analyses: Analyses of stable and radiogenic isotopes are of high value to investigate the long-term integrity of salt formations. It is possible to show, from isotopic analysis of fluid inclusions, brines and embedded minerals, that some interbedded salt/clay horizons have remain unchanged since the time of their deposition. This provides evidence that, under certain conditions, rock salt may be unaffected by intrusion of external groundwaters over very long geological time scales.
- The viability of microbes in the near-field: It is accepted that microbes will be present but unclear whether they are significant. Information from old mines, operational disposal sites (WIPP, Morsleben, etc.) might be useful to determine whether populations are viable and could actually impact the integrity of the repository, e.g. by contributing to gas generation or formation of corrosive substances to damage the barriers.
- The significance of disseminated hydrocarbons: A repository will be sited away from large accumulations of oil and gas, but hydrocarbons may be finely disseminated throughout rock salt. Similar to the issue of microbes, information from deep old mines or from cores of deep boreholes (deeply buried salt formations), etc., might be useful to determine whether thermochemical reduction of sulphates is an observed phenomenon on a large scale, so could be an actual risk to the repository, or only a theoretical possibility.
- Material degradation and corrosion: Different materials will be used in the
  construction of waste containers and the engineered barriers, and some of these
  may degrade or corrode in the highly saline near-field environment. Not all modern
  materials have an archaeological analogue, but examination of artefacts from old
  mines could provide bounding limits on what materials degrade and which are
  stable in both dry and humid saline environments.

#### References

- Hammer, J., et al. (2012), "Salzgeologische Bewertung des Einflusses von "kryogenen Klüften" und halokinetischen Deformationsprozessen auf die Integrität der geologischen Barriere des Salzstocks Gorleben", Bericht zum Arbeitspaket 2, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-273, ISBN: 978-3-939355-49-6, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Köln.
- Miller, W.M., et al. (2006), Network to Review Natural Analogue Studies and Their Applications to Repository Safety Assessment and Public Communication (NAnet). Synthesis Report, European Commission, EUR Report 21919.
- Miller, W.M. and L.A. Kovach (1993), The Role of Natural Analogs in Geologic Disposal of High-Level Nuclear Waste, Nuclear Regulatory Commission (NRC) Report, CNWRA-92-020.
- Noseck, U., et al. (2008a), "Identification of Uranium Enrichment Scenarios by Multi-Method Characterisation of Immobile Uranium Phases", J. Phys. Chem. Earth, 33, 969-977.
- Noseck, U., I. Müller-Lyda, W. Steininger (2008b), "Overview on Natural Analogue Studies in Germany", atw, 53, 342-348.

#### NEA/RWM/R(2013)10

- Noseck, U., et al. (2009), "Carbon Chemistry and Groundwater Dynamics at Natural Analogue Site Ruprechtov, Czech Republic: Insights from Environmental Isotopes", Applied Geochemistry, 24, 1765-1776.
- Organisation for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) (2000), Regulatory Reviews of Assessments of Deep Geologic Repositories: Lessons Learnt, NEA Radioactive Waste Management Report, OECD/NEA, Paris.
- OECD/NEA (2012), Methods for Safety Assessment of Geological Disposal Facilities for Radioactive Waste: Outcomes of the NEA MeSA Initiative, NEA Radioactive Waste Management Report, No. 6923, OECD/NEA, Paris.
- Wolf, J., U. Noseck, A. Meleshyn (2013), Anwendbarkeit von Natürlichen Analoga für den Langzeitsicherheitsnachweis für ein Endlager im Salzgestein, GRS Report, forthcoming.

# Session I General Aspects of Analogues

#### NEA/RWM/R(2013)10

## The process of defining an optimal natural analogue programme to support national disposal programmes

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Natural analogue studies are often criticised for missing their target and failing to provide appropriate data for repository safety assessment. It is true that, in many cases, analogues have been oversold and ended up more academic exercises than contributions to the safety case. However, others have provided invaluable input that cannot be obtained from any other source. The key to success seems to require that national NA programmes be appropriately formulated, using a top-down (rather than the standard bottom-up) approach. Although in the past depending on input from "gurus" with wide experience in both safety assessment and natural analogues, this can now be developed by a formal methodology which borrows from recent developments in the field of knowledge management. This is applicable to any national programme and has already led to the development of a specific NA study which is currently ongoing in the Philippines.

#### **Background**

The field of "natural analogues" (NA) developed predominantly in a bottom-up manner, led predominantly by geoscientists (and a few archaeologists) who recognised that extrapolations over long time scales were one of the major weaknesses of safety assessments for radioactive waste disposal – especially for longer-lived waste. Many early analogues focused on material properties, where extrapolations from laboratory studies to geological time scales were particularly weak. Nevertheless, the wider applications for "validating" (or testing) models and databases and also presenting key arguments to non-technical audiences were recognised and led to rapid expansion of the field (described in several overviews – e.g. McKinley, 1998; Miller, et al., 2000; West, et al., 2002). The problem that rapidly emerged was that many of the studies initiated were poorly conceived, planned and implemented by teams with little understanding of the details of the safety assessments in which output would be used and, despite such limitations, were grossly oversold. Even simple PR applications were spoiled by lack of technical understanding and blatant hyperbole.

Despite such problems, the attraction of NAs remains clear and, indeed, the requirement to provide supporting analogue arguments to complement traditional performance assessments is now often enshrined in national legislation and is integral within the concept of a "safety case". The question is thus – how can we learn from the positive and negative experience in the past and develop new national NA programmes that live up to their promise? This was the aim of a project initiated by RWMC (Japan) almost ten years ago (McKinley and Alexander, 2007a,b; Alexander, et al., 2007), which led to a NA study currently running in the Philippines (e.g. Alexander, et al., 2008; McKinley,

et al., 2008). The approach developed to plan an optimised national programme is, however, more widely applicable and can be extended to the specific priorities that have developed in recent years.

#### An optimised approach to developing a national NA programme

The process involved in deriving the requirements for an optimised NA programme should be strictly structured, but this has rarely been the case in the past. In the few cases when NAs have been sought to match a particular end user need, this requirement has generally been identified by experienced individuals with a general or NA co-ordination role in national R&D programmes. The process involved can be more formally structured by a bottom-up critical assessment of openly-available NA literature, with particular emphasis of its relevance to specific national geological disposal programmes. With an emphasis on technical aspects, focus is placed on the requirements to provide input to the development of safety cases for such national programmes.

This was carried out by RWMC, starting from an assessment of the weightings given to the wider aims of the NA programme, taking into account secondary aims associated with the "safety strategy" and the practical constraints resulting from the likely duration of the NA project when compared to national programme deadlines. This then leads to specification of optimised NA project(s) in a top-down manner, representing main goals in terms of individual work packages (WPs), bearing in mind that there can be synergies involved in combining such WPs at a suitable location. This concludes with a process for site selection, which can also be carried out in a structured way using some form of multi-attribute analysis (MAA – cf. Alexander and Milodowski, 2008). The RWMC procedure is outlined in more detail below to form the basis for consideration of how it could be widened to be more relevant to other national programmes.

#### Bottom-up development of a preliminary NA "wish list"

This process was initiated by a critical review of the NA literature, producing summaries of key publications and producing an archive of all relevant material. This provides input to the process, in terms of identification of areas where required input for the Japanese programme can be obtained from existing published work and other relevant areas where little information is available. This is the knowledge base that forms the core around which safety case support will be built.

Critical review of everything available in the analogue field allowed quality levels to be assigned to projects and publications. This led to more detailed assessment of a few "key" NA projects, allowing further assessment of the relevance of output for the Japanese national programmes and the potential of extensions of such work to fill gaps in available information. This was complemented by a superficial overview of the requirements for a safety case based on the procedure outlined elsewhere – in this case, JNC's H17 report (JAEA, 2005) was used as an example (deep geological disposal of HLW, with the option of considering co-disposal of TRU).

The assessment of the requirements for making the safety case is outlined below, based on expert opinion on the likely level of information available at the time of site selection. It should be emphasised that, as implied by the bottom sub-divided box in Figure 1, the distinction between generic and site-specific work is often artificial and, indeed, this will tend to become even more blurred with time, as the project moves on to the full site characterisation stage.

The individual areas where NA studies can be used can be listed, using the numbering system used in H17, and are shown in the figure. It should be emphasised, however, that this representation of making the safety case does not cover all key applications of

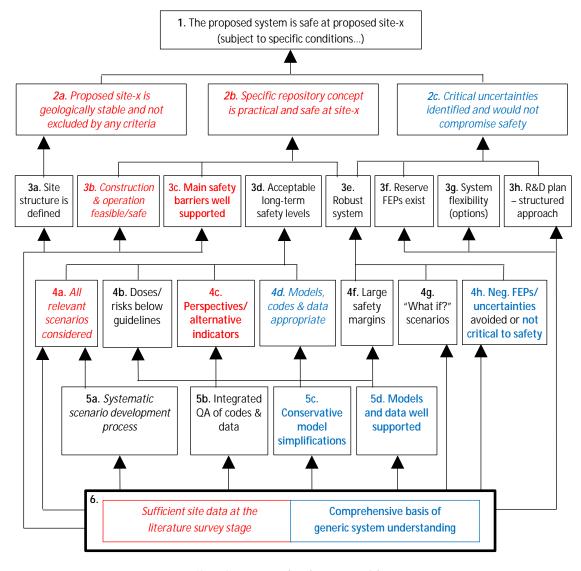


Figure 1: Flow of information to make the safety case for the example of a deep geological repository in Japan (H-17) at the PIA stage

Key – Assessment of analogue potential:

Blue – generic, Red – dependent on site selected, Black – none, Bold – important role, Italic – minor role

NAs, in particular the "higher level" goals included in the safety strategy (e.g. establishment and maintenance of an experienced work force) and also special requirements for presenting the safety case to key stakeholders, which will be considered at the top level of establishing project aims as described below.

It is also important to note that higher level boxes in this flow chart would tend to be supported by system analogues, while lower levels are supported by sub-system or process analogues. It should further be emphasised that, for technical applications, NA studies are rarely very useful in isolation, but are most effective when integrated in a combined programme of laboratory, modelling and in situ studies (cf. Alexander, et al., 1998).

For training purposes, complex, integrated NAs – which study systems or sub-systems (e.g. Poços de Caldas, Cigar Lake) – are generally best for accumulation of interdisciplinary

experience, which is difficult to obtain elsewhere. For stakeholder communication, system NAs are often used (e.g. Oklo), but there is a great danger of over-interpretation of the analogy due to the simplifications that have to be introduced (see discussion in McKinley and Alexander, 2007a; Alexander, et al., 2007). For unambiguous communication, simple sub-system analogues (e.g. materials longevity) are usually best.

Looking more closely at Figure 1, several types of NA study could be identified, which could be appropriate in supporting certain aspects of the safety case.

#### Generic analogue studies

- 2c. Critical uncertainties identified and would not compromise safety: System analogues –
  no additional processes identified which are not included in the analysis (weak use
  of analogues, absence of evidence rather than evidence of absence hence should
  not be over-interpreted: see discussion in NAWG, 1995; Miller, et al., 2000).
- 4d. Models, codes and data appropriate: Scenario-specific analogues; generally weak application for complex systems (ability to simulate) rather than rigorous testing (blind model prediction; cf. Pate, et al., 1994).
- 4g. Negative FEPs/uncertainties avoided or not critical to safety: Sub-system analogues focused on potential problem areas, e.g. gas, colloids, microbes, organics (e.g. McKinley and Grogan, 1988).
- 5c. Conservative model simplifications: All models involve simplifications of the real system; focused analogue studies can determine if these are truly conservative for relevant systems, e.g. assumed conservatism of Kd sorption model (McKinley and Alexander, 1992).
- 5d. Models and data well supported: Key role of specifically testing PA-relevant models and databases in relevant environments/over relevant time scales, e.g. material degradation, radionuclide release and transport (e.g. McKinley, et al., 1998).
- 6. Comprehensive basis of generic system understanding: Especially critical for extending understanding over long time periods but also includes generic understanding of the properties of undisturbed geological systems (especially relevant to radionuclide transport; e.g. Noseck, et al., 2012).

#### Site-specific (siting environment type) analogues

- 2a. The proposed site-X is geologically stable and not excluded by any criteria: Studies carried out in environments analogous to that of the SITE [NB not within usual definitions of analogues, cf. Nagra's use of the Mont Terri site data in the Opalinus Clay safety case (Nagra, 2002a)].
- 2b. Specific repository concept is practical and safe at site-x: Complete system analogues, e.g. ore body in coastal location for a coastal SITE (cf. Smellie and Karlsson, 1996).
- 3b. Construction and operation feasible/safe: Industrial analogues (mining, underground civil engineering), generally not included in usual analogue definition, but see examples of their use in Posiva (2012).
- 3c. Main safety barriers well supported: Arguments from analogues to support assumed performance of barriers at the proposed site, e.g. support for assumed bentonite properties under conditions with varying groundwater chemistry (fresh « marine; Posiva, 2012).
- 4a. All relevant scenarios considered: Analogues of safety barriers in relevant siting environments, with special consideration of time-varying processes, e.g. marine transgressions/regressions in coastal areas (e.g. Smellie and Karlsson, 1996).

- 4c. Perspectives/alternative indicators: Natural fluxes of radionuclides, etc.; could include biosphere analogues (cf. Neall and Smith, 2001).
- 6. Sufficient site data at the literature survey stage: NB palaeohydrogeology often included with analogues (cf. Nagra, 2002b and discussions in NAWG, 1995).

#### Synthesis

The special areas identified as potential topics for future NA studies can be grouped as:

- Settings or host rocks which are little studied elsewhere:
- active tectonic situations;
- salt host rock;
- small islands.
- Special areas of study:
  - palaeohydrogeology;
  - radionuclide (solute) transport processes (from ore body or geochemical anomaly);
  - processes of fault reactivation and self-sealing;
  - effects of perturbations colloids, organics, microbes, gas;
  - construction/operation (engineering; expected evolution, perturbations).
- Process understanding:
  - saltcrete-bentonite/host-rock interaction;
  - radionuclide migration within the EBS;
  - model/database testing;
  - radionuclide solubility/speciation within overpack;
  - realistic steel corrosion model (rate/gas production);
  - retardation of radionuclides on anaerobic corrosion products;
  - retardation of radionuclides on saltcrete alteration products;
  - long-term behaviour of salt at higher repository temperatures.
- · Uncertainties and perturbations:
  - rheology of salt (long-term waste tunnel movement);
  - long-term performance of shaft and tunnel seals;
  - role of microbes in the near-field.

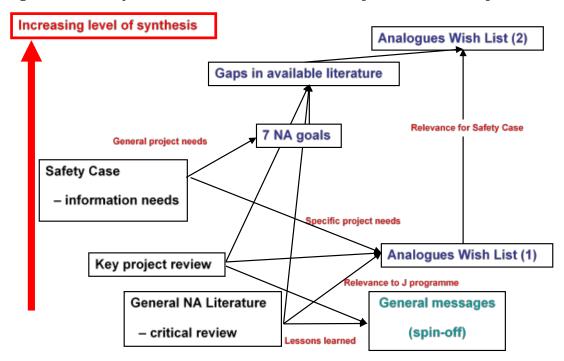
The input of literature surveys (also called "data mining") could be integrated with the identified needs above to derive a preliminary "wish list" of possible NA projects. This list of potential projects also included a very rough, subjective assessment of a wide range of issues of relevance to any national programme overall and the safety case in particular:

- technical complexity;
- potential for model testing;
- · possibility of production of repository relevant data;
- · risk of failure (i.e. desired output not achievable within budget and time constraints);
- · cost:

- · potential for staff training;
- extent of application for stakeholder communication purposes;
- potential international interest in collaboration in the project;
- · minimum duration (under ideal conditions).

In summary, the flow of information to produce this "NAs Wish List (1)" is presented in the bottom half of Figure 2. It should be noted that, in addition to the documented reviews, some general messages on the applicability of NA literature were produced as a form of spin-off from the work.

Figure 2: Summary of information flow in the bottom-up assessment of requirements



#### **Derivation of safety goals**

In addition to the specific project requirements listed above, the general areas of the safety case where NAs could contribute can be reformulated as a series of goals – as also indicated in Figure 2. These are considered top-down in terms of safety case hierarchy, with sub-divisions in terms of aspects which are either waste- or site-specific.

#### Goal 1: Demonstration of geological stability

The assurance of geological stability is sine qua non for a deep disposal project and the site-specific database may be rather limited and hence it may be useful to complement this work by studies of similar rocks in similar geological settings, where the ability to access appropriate information may be greater. Such studies can be classified as focusing on a "self analogue", although this is not common terminology (e.g. such arguments were the basis of establishing the Grimsel and Mont Terri URLs in Switzerland, but these were called "first generation" URLs rather than NAs). The type of NA study which could be envisioned is further subdivided as involving assessment of the:

stability of host rock;

- stability of setting;
- · robustness to perturbations.

These topics are closely related and result from the fact that the present characteristics of any potential host rock result from its original properties at the time of its formation, modified by the perturbations accumulated during its history. In the case where the host rock/setting has been disturbed, there may be processes which minimise the effects on key geological characteristics – e.g. plastic deformation or self-healing of a salt or soft clay host rock. In general, the less a rock has been altered over geological time and the more stable its geological setting has been, then the easier it will be to characterise and to predict its future evolution. Examples of the type of work that could be carried at NA sites would be:

- assessment of stability of overlying sediment in the case of deep burial and subsequent uplift by examining mineral transformations;
- assessment of the effects of local folding on rock mechanical and hydrogeological properties;
- · quantification of self-healing of fractures and faults.

Such information could, for example, be obtained from tunnels or other underground constructions outside the actual site (cf. Nagra, 2002a,b). Note that this work is generally considered part of site characterisation in most national programmes and hence terminology should be clarified before initiating such work in any national programme.

#### Goal 2: Demonstration of deep groundwater stability

As groundwater release scenarios are the major concern in post-closure safety assessment, the stability of the hydrogeological environment is also a key concern. This is subdivided into:

- · flow stability;
- · chemical stability.

It is generally desirable that the groundwater in the host rock and surrounding formations should be characterised by both low fluxes and low velocities. As these parameters are often difficult to measure directly, a useful surrogate measure is groundwater age – very old groundwater being an indirect indication that both fluxes and velocities are low (see discussion in Waber, et al., 2012, for example). The determination of the history of site hydrology – palaeohydrogeology – is an established discipline and part of normal site characterisation (e.g. Ota, et al., 2010). The particular interest here would be studying hydrogeology at self-analogue sites (as considered above) when these would provide better access than the proposed repository site (cf. Niizato, et al., 2010).

In addition to the stability of hydrogeology, hydrochemical stability is also important in determining the performance of both the engineered and the natural barriers. Preferred hydrochemistry would include high salinity, chemically reducing conditions in the host rock and neutral – mildly alkaline pH and low salinity – and chemically reducing conditions in the surrounding formations. NA studies could examine the extent to which such conditions were buffered by the host rock, particularly under any major perturbations that may occur at the site of interest over relevant time periods (e.g. marine transgression/regression, flushing with oxidising water as a result of de-glaciation (e.g. Noseck, et al., 2009).

As noted above, in most national programmes, such work is considered part of site characterisation and not classified as being within the remit of NAs, but excluding this from such a holistic assessment risks missing the potential support to the safety case that such studies can offer.

#### Goal 3: Demonstration of EBS robustness

Particularly at early stages of the siting process, repository relevant geological databases are likely to be rather uncertain and hence the safety case is likely to focus on the behaviour of the EBS. Particularly important in this case is the robustness of EBS performance, even in the case of minor perturbations or degradation of the natural barrier. The robust performance of the EBS results from the accumulated barrier roles of materials and processes, including:

- · steel;
- bentonite/clay;
- · cementitious materials;
- · redox buffering (including microbes);
- · radionuclide solubility;
- · radionuclide transport (including colloids,...);
- · gas (production, release,...).

Study of such materials and processes has been a past focus for NA studies and a significant volume of relevant information exists in the international literature. Although it is important to assess the relevance of individual studies to the repository project conditions in any specific country, much of the EBS work is fairly generic and so could be profitably data mined. A good recent example of this is the work on potential grout leachate reaction with bentonite in the Swedish programme (Sidborn, et al., 2012).

### Goal 4: Demonstration of natural barrier performance

The natural barrier role is a key aspect of any site and hence information on this may help to support decisions associated with preliminary site selection. The aim would be to look for evidence, at either the proposed site or one which can be considered analogous, which would allow conclusions to be drawn about key processes such as:

- · radionuclide sorption;
- · matrix diffusion;
- · dilution and dispersion;
- radionuclide immobilisation (trapping);
- · colloid effects (including organics);
- · microbes:
- · gas.

As considered above, such studies have been a past focus of NA studies and a considerable literature exists of potentially relevant work. In this case, however, applicability is very much dependant on the host rock and siting environment.

### Goal 5: Demonstration of robustness to engineering

In the many national repository concept development processes, the impact of construction, operation and closure of a repository on its long-term performance is identified as an issue to be considered at early stages. This would be particularly important in any case when long-term access for institutional control is considered. The aspects of interest are associated with the response of a rock in a particular setting to construction and operation, especially:

- · recovery of hydrology;
- · recovery of geochemistry;
- · recovery of rock mechanics.

Such processes could be studied at construction sites, mines, etc., either in the proposed siting region or somewhere similar. Again, such studies are not generally termed NAs when carried out in most national programmes (often part of engineering geology site characterisation), but their great potential should not be ignored (see Posiva, 2012, for excellent examples).

#### Goal 6: Demonstration of robustness to site evolution

The robustness of particular sites to the perturbations which may be expected to occur as part of long-term evolution is clearly an attribute which is important to consider when making a site selection. This has recently been receiving more interest for coastal locations in particular (e.g. Milodowski, et al., 2005; Salas, et al., 2010; Posiva, 2012), due to concerns about the effects of sea level change, but the approach is completely valid for any potential site (cf. Alexander, 2010). A few examples of the topics which could be relevant would be:

- · glacial transitions;
- · climatic biosphere cycles;
- coastal sea-level changes;
- · GBI alteration (erosion/sedimentation, transgression/regression, etc.).

Although little studied directly in the past, NA methods would be applicable to interpret the consequences of past processes of this type at a proposed site (or a similar location as noted above) in order to estimate what might be likely to occur in the future. Indeed, it has been noted that concerns about site robustness in the broadest sense are often raised by non-technical stakeholders (see, for example, West, et al., 2002; NDA, 2012).

## Goal 7: Demonstration of insensitivity to EBS component interactions

Past generations of SA models have tended to simplify the treatment of EBS components, treating them as effectively independent of each other. In order to develop a safety case, however, such assumptions need to be shown to be valid. The interactions which may be important include:

- · steel waste:
- bentonite steel;
- · bentonite cementitious materials.

As with Goal 3 above, these reflect the types of study which have been included in past analogue projects (e.g. Mitsui, et al., 1996a,b) – although interactions have certainly been looked at less rigorously than the behaviour of materials in isolation (but see Milodowski, et al., 2002 for a good example).

## Overview: Top-down establishment of an optimised NA project

The example here illustrates a top-down approach to defining and developing relevant NA studies applied in Japan. Of note is the fact that this method has led to the establishment of a highly successful and novel study: the Philippines Alkaline Natural Analogue Study, where the potential reaction of industrial bentonite with alkaline leachates from low-alkali cements is being studied at the Zambales ophiolite in Luzon (see Alexander, et al., 2008; McKinley, et al., 2008; Arcilla, et al., 2009; Fujita, et al., 2010 for

details). In principle, however, the same structured approach can be applied for the boundary conditions of any national programme.

## Extension of approach to other programmes

As a "quick and dirty" example, the approach outlined has been informally applied to the specific requirements for HLW/SF disposal in salt as considered in the German national programme. Using the logic outlined in Figures 1 and 2, the generic goals can be compared with information available in the literature, resulting in the overview shown in Table 1 for most sites world wide, with a few specific examples included for a salt host rock. The goals can also be mapped against the original NA project wish list, which can allow priorities to be identified as illustrated with a few examples in Table 2. As noted above, there are a number of complementary aims which can be considered when devising an optimised NA project. These would include:

- increasing/confirming system understanding for new siting environments/host rocks;
- developing understanding of novel repository concepts;
- testing of new models/databases (validation);
- · training new staff and facilitating know-how transfer.

The approach outlined allows these to be assessed in a structured manner to produce a programme tailored to specific national needs – or, indeed, the needs of a group of countries if they have similar boundary conditions and would like to collaborate in this field. Since the initial RWMC study, Japanese work on advanced knowledge management systems has advanced considerably and these could be readily incorporated into the processes of both developing consensus on the weighting of project options (e.g. using argumentation modelling) and also communicating results – both between the multidisciplinary members of a project and to a wider audience of stakeholders.

## Table 1: Example of potential NA studies which map to the aims of the goals noted above

	Potential NA studies						
Major site		Minor site	Archaeological	Process	Comments		
Goal 1: Demonstration	NB very site specific						
Stability of host rock	Gas storage caverns in salt (Europe wide)	WIPP					
2. Stability of setting	Gas storage caverns in salt (world wide)	North Sea and northern Germany in general			Most international work not in the "analogue" literature		
3. Robustness to perturbations	Lisbon Valley, Utah	North Sea gas caps			Most international work not in the "analogue" literature		
Goal 2: Demonstration	Goal 2: Demonstration of deep groundwater stability						
1. Flow stability	WIPP	Mont Terri (diffusive)		Mont Terri (diffusive)	NB very site specific; international work irrelevant		
2. Chemical stability Mor		Mont Terri			Most international work not in the "analogue" literature		
Goal 4: Demonstration	on of natural barrier	performance (pa	rticularly in overlyir	ng rock			
1. RN sorption	Ruprechtov	Loch Lomond	och Lomond Var		Many attempts in literature, but applicability in most cases debatable		
2. Matrix diffusion	Poços de Caldas	Grimsel	Akrotiri	Various	Lots of data available		
3. Dilution & dispersion	Ruprechtov	Needle's Eye, Broubster, Alligator Rivers			Very difficult to disentangle from other processes		
4. RN immobilisation (trapping)	Ruprechtov, Poços de Caldas, Oklo, Cigar Lake, Loch Lomond	Needle's Eye, Broubster		Various	Much empirical information but mechanisms not well resolved		
5. Colloid effects (incl. organics)	Gorleben	Morro do Ferro, Cigar Lake		Various	Many studies show colloids present but little demonstration of significance		

	Irrelevant
	Some relevance
	Fair amount of relevance
	Topic of potentially high importance

Table 2: Examples of mapping a NA "wish list" to the programme goals

Site	Aim	Goal 1	Goal 2	Goal 3	Goal 4	Goal 5	Goal 6	Goal 7	Comment
Ruprechtov	Radionuclide retardation								Topic relevant and similar environment
European gas storage sites	2. Fault healing								Process relevant, but the tectonic setting needs careful assessment
Lisbon Valley	3. Ore body microbiology								Redox traps, immobilisation; could be important for a site needing a powerful EBS
	4. Radio- chemistry								Potential to test models of key retardation process in near- and far-field
	5. Redox traps								Redox traps, immobilisation; could be important for a site needing a powerful EBS
	6. Colloids								Potential to test models of potential NBS short circuit in a well-defined flow system (although chemistry may not be so relevant)

Irrelevant
Some relevance
Fair amount of relevance
Topic of potentially high importance

## References

Alexander, W.R. (2010), "The Impact of a Hyperalkaline Plume on Fractured Crystalline Rock", Proc. NEA-IGSC Workshop on Cementitious Materials in Safety Cases for Geological Repositories for Radioactive Waste: Role, Evolution and Interaction, Brussels, Belgium, 17-20 November 2009, OECD/NEA, Paris, France.

Alexander, W.R. and I.G. McKinley (1992), "A Review of the Application of Natural Analogues in Performance Assessment: Improving Models of Radionuclide Transport in Groundwaters", J. Geochem. Explor., 46, pp. 83-115.

Alexander, W.R. and A.E. Milodowski (2008), Low-Alkali Cement Leachate/Bentonite Interaction Natural Analogue Study, Cyprus Natural Analogue Project (CNAP) Phase I. Bedrock Geosciences Technical Report BG08-01, Bedrock Geosciences, Auenstein, Switzerland.

- Alexander, W.R., A. Gautschi and P. Zuidema (1998), "Thorough Testing of Performance Assessment Models: The Necessary Integration of In Situ Experiments, Natural Analogues and Laboratory Work", Extended Abstract in Sci. Basis Nucl. Waste Manag., XXI, 1013-1014.
- Alexander, W.R., I.G. McKinley and A.B. MacKenzie (2007), Development of an Optimised Natural Analogue Programme to Support the Japanese HLW/TRU Disposal Programme (Part 1), MCM-TR-07-01, MCM, Baden-Dättwil, Switzerland.
- Alexander, W.R., et al. (2008), "A New Natural Analogue Study of the Interaction of Low-Alkali Cement Leachates and the Bentonite Buffer", Sci. Basis Nucl. Waste Manag., XXXI, 493-500.
- Arcilla, C.A., et al. (2009), "Reaction Pathways for Rising Hyperalkaline Groundwater in a Bentonite Mine in the Philippines", Proc. Goldschmidt 2009 Conference, Davos, Switzerland, 22-26 June 2009, Geochim. Cosmochim. Acta, Goldschmidt Conference Abstracts, A50.
- Fujita, K., et al. (2010), "Natural Analogue Study of the Interaction Between Hyperalkaline Groundwater and Bentonite at the Ophiolite of Philippine", Proc. Japan Geoscience Union Meeting, Makuhari, Japan, 23-28 May 2010, JGU, Tokyo, Japan.
- Japan Atomic Energy Agency (JAEA) (2005), H17: Development and Management of the Technical Knowledge Base for the Geological Disposal of HLW, Knowledge Management Report, JNC TN1400 2005-022, JAEA, Tokai, Japan.
- McKinley, I.G. (1989), "Applying Natural Analogues in Predictive Performance Assessment (1): Principles and Requirements; (2): Examples and Discussions", Risk Analysis in Nuclear Waste Management, Klewer Academic Publ., the Netherlands, pp. 357-396.
- McKinley, I.G. and H.A. Grogan (1986), "Natural Analogue Support for Geomicrobiological Modelling", in B. Côme and N.A. Chapman (Eds.), CEC Nuclear Science and Technology Report, EUR 10671, Natural Analogue Working Group, Second Meeting, Interlaken, June 1986, 153-159, CEC, Luxembourg.
- McKinley, I.G. and W.R. Alexander (1992), "Constraints on the Applicability of In Situ Distribution Coefficient Values", J. Environ. Rad., 15, pp. 19-34.
- McKinley, I.G. and W.R. Alexander (2007a), Development of an Optimised Natural Analogue Programme to Support the Japanese HLW/TRU Disposal Programme (Part 2), MCM-TR-07-02, MCM, Baden-Dättwil, Switzerland.
- McKinley, I.G. and W.R. Alexander (2007b), Development of an Optimised Natural Analogue Programme to Support the Japanese HLW/TRU Disposal Programme (Obayashi Work Components), MB-TR-07-01, McKinley Consulting/Bedrock Geosciences Report, Baden-Dättwil, Switzerland.
- McKinley, I.G., et al. (2008), "IPHAP: A New Natural Analogue of Bentonite Alteration by Cement Leachates", Proc. Int. Symp. Radiation Safety Management, Daejeon, Korea, 7-9 November 2007, pp. 492-498.
- Miller, W.M., et al. (2000), "Geological Disposal of Radioactive Wastes and Natural Analogues", Waste Management Series, Vol. 2, Pergamon, Amsterdam, the Netherlands.
- Milodowski, A.E., et al. (2002), Alteration of Uraniferous and Native Copper Concretions in the Permian Mudrocks of South Devon, United Kingdom, SKB Technical Report, TR-02-09, SKB. Stockholm. Sweden.
- Milodowski, A.E., E-L. Tullborg, et al. (2005), Application of Mineralogical, Petrological and Geochemical Tools for Evaluating the Palaeohydrogeological Evolution of the PADAMOT Study Sites, PADAMOT Project Technical Report WP2, 228 pp, EU 5<sup>th</sup> Framework Programme, Contract No. FIKW-CT-2001-00129, EU, Luxembourg.

- Mitsui, S., M. Kubota and R. Murakami (1996a), "An Investigation of Environmental Conditions of Soil Around a Buried Bronze Bell from the Ruins of Shimoda", Res. Rep. Osaka Centre Cultural Heritage, 18, pp. 360-370.
- Mitsui, S., M. Kubota and R. Murakami (1996b), "Burial Conditions of a Bell-Shaped Bronze Artifact Excavated at the Shimoda Site", Res. Rep. Osaka Centre Cultural Heritage, 18, pp. 371-381.
- National Cooperative for the Disposal of Radioactive Waste (Nagra) (2002a), Demonstration of Disposal Feasibility for Spent Fuel, Vitrified High-Level Waste and Long-Lived Intermediate-Level Waste (Entsorgungsnachweis), Nagra Technical Report NTB 02-05, Nagra, Wettingen, Switzerland.
- Nagra (2002b), Projekt Opalinuston: Synthese der geowissenschaftlichen Untersuchungsergebnisse – Entsorgungsnachweis für abgebrannte Brennelemente, verglaste hochaktive sowie langlebige mittelaktive Abfälle, Nagra Technical Report NTB 02-03, Nagra, Wettingen, Switzerland (in German).
- Natural Analogue Working Group (NAWG) (1995), Proc. 6<sup>th</sup> NAWG Workshop, Santa Fe, NM, 12-16 September 1994, CEC EUR Report EN16761.
- Nuclear Decommissioning Authority (NDA) (2012), Geological Disposal: RWMD Approach to Issues Management, NDA Report No. NDA/RWMD/081 Version 2, March 2012, NDA-RWMD, Harwell, UK.
- Neall, F.B. and P.A. Smith (Eds.) (2001), H12: Examination of Safety Assessment Aims, Procedures and Results from a Wider Perspective, JNC Technical Report JNC TY1400 2004-001, JAEA, Tokai, Japan.
- Niizato, T., et al. (2010), "Development of Comprehensive Techniques for Coastal Site Characterisation: (3) Conceptualisation of Long-Term Geosphere Evolution", Proc. ICEM 2010 Conference, ASME, Washington, USA.
- Noseck, U., K. Rozanski, et al. (2009), "Carbon Chemistry and Groundwater Dynamics at Natural Analogue Site Ruprechtov, Czech Republic: Insights from Environmental Isotopes", Appl. Geochem., 24, 1765-1776.
- Noseck, U., E-L. Tullborg, et al. (2012), "Real System Analyses/Natural Analogues", Appl. Geochem., 27, 490-500.
- Ota, K., et al. (2010), "Development of Comprehensive Techniques for Coastal Site Characterisation (1) Strategic Overview", Proc. ICEM 2010 Conference, ASME, Washington, USA.
- Pate, S.M., I.G. McKinley and W.R. Alexander (1994), Use of Natural Analogue Test Cases to Evaluate a New Performance Assessment TDB, CEC Report EUR15176EN, Luxembourg.
- Posiva (2012), Safety Case-2012: Complementary Considerations, Posiva Report 2012-11, Posiva, Eurajoki, Finland (forthcoming).
- Salas, J., et al. (2010), SR-Site Hydrogeochemical Evolution of the Forsmark Site, SKB TR10-58, SKB, Stockholm, Sweden.
- Sidborn, M., N. Marsic, et al. (2012), Potential Alkaline Conditions for Deposition Holes of a Repository in Forsmark as a Consequence of OPC Grouting, SKB TR 2012-XX, SKB, Stockholm, Sweden (forthcoming).
- Smellie, J.A.T. and F. Karlsson (Eds.) (1996), The Cigar Lake Analogue Project: A Reappraisal of Some Key Issues and Their Relevance to Repository Performance Assessment, SKB Technical Report, TR 96-08, SKB, Stockholm, Sweden.

- West, J., et al. (2002), "Communication of Nuclear Power Concepts to Stakeholders The Use of Nature's Own Laboratories", Proceedings of NUCEF 2001 Scientific Bases for Criticality Safety, Separation Process and Waste Disposal, Tokai, Japan, 31 October-2 November 2001, Japan Atomic Energy Research Institute Report, JAERI Conf. 2002-004. pp. 47-54, JAEA, Tokai, Japan.
- Waber, H.N., T. Gimmi and J.A.T. Smellie (2012), "Reconstruction of Palaeoinfiltration During the Holocene Using Porewater Data (Laxemar, Sweden)", Geochim. Cosmochim. Acta, 94, 109-127.

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## Analogues, analogies, anecdotes and more – Findings of the NEA Forum on Stakeholder Confidence

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The use of analogues to help understand and to build confidence in radioactive waste management approaches and safety cases was reviewed by the NEA Forum on Stakeholder Confidence. Analogues can only contribute to safety cases as a line of evidence. They cannot be relied upon as the sole line of evidence. As the degree of similarity with possible repository situations diminishes, examples become analogies or anecdotes, and their arguments are more useful in terms of supporting generic and conceptual feasibility of geological disposal. Analogies and anecdotes could help the public to grasp time scale and understand the basic rationale and principles of geological disposal. This is more important in early stages of public information and involvement. When the project becomes more real, however, the public may find a national analogue more persuasive and comprehensible, since it is geographically and culturally closer to its own experience and likely to be very concrete, especially in the case of anthropogenic analogues. The application of contemporary analogues as input to the design and choice of materials with a view to suggest safe and reversible operation deserves attention.

## Introduction

As part of its programme of work the OECD/NEA Forum on Stakeholder Confidence¹ continues to investigate the theme of the "Link Between RD&D and Stakeholder Confidence".

Regulators need a technical demonstration to aid in evaluating the safety case. Local stakeholders appreciate the opportunity to visualise technological arrangements. In both cases, demonstration adds to confidence in the feasibility of solutions. Some believe that, if handled with integrity, there is an important role for analogues in both the technical safety case and in communication with stakeholders.

A topical session was held on 4 June 2008 on the use of analogues to help understand and to build confidence in radioactive waste management approaches and safety cases. Case studies were presented from Finland, Spain and Switzerland and from joint international endeavours, namely the EC projects NANET and PAMINA. The results of a questionnaire filled by FSC members in co-ordination with their IGSC colleagues served as the basis of the introductory presentation.

This paper presents the main results of the 2008 FSC topical session, which is also documented on the FSC open web page (NEA, 2008).

<sup>1.</sup> www.oecd-nea.org/rwm/fsc.

#### **Discussion**

## FSC calls attention to the proper use of the word "analogue"

According to the degree of similarity to the repository and its environment, the situation or example in question may be considered to be an analogue, an analogy or an anecdote. The FSC used the following definitions to frame its observations.

When a fairly direct similarity to repository situations exists, the case may be used as an argument to support a phenomenological theory and its modelling: it is an analogue. As the degree of similarity decreases (e.g. if relevant chemical or physical conditions do not apply with those of the repository and the consequences of these deviations cannot be quantified; if system boundaries cannot be defined), the case applies more narrowly to the generic feasibility of medium- to long-term safety of geological disposal. When the degree of similarity is quite low, the case may provide a "common sense" rationale supporting the concept of geological disposal as an option that should not be ruled out and could be a relevant solution, providing that adequate research and demonstration are performed to confirm this hypothesis. In the latter cases, the situation is considered an analogy or even only an anecdote. The following example illustrates the FSC definitions of analogue, analogy and anecdote:

- · The case of Roman nails found in Scotland can be presented as:
  - An analogue for the analysis of corrosion resistance, were it decided to use steels that are viewed to be as corrosion-resistant as or more corrosion-resistant than the metal of which the nails were made. Indeed, the evidence of their longevity would contribute to modelling confirmation, despite the related uncertainties.
  - An analogy to illustrate the confinement properties of natural clay over a long time period, as well as their ability to reduce corrosion. We cannot go beyond analogy here because the initial number of nails buried is unknown (we cannot state how many nails corroded away).
  - An anecdote, simply to show that man-made artefacts can last thousands of years underground, if it were used by a programme that is not contemplating the use of steel for containers or clays as a barrier.

## Why use analogues?

Time scales relevant to long-term safety of geological disposal (on the order of several centuries, millennia and sometimes more) cannot be attained in experiments. Analogues can provide qualitative or quantitative data in order to illustrate long-term behaviour or conditions and to validate assessment models. They can also provide references and examples, and confirm phenomenological forecasts over long time scales.

Important limitations include, however, that the initial and evolution conditions cannot always be well known and that the materials used may be different from the ones to be considered in a repository. Also, analogues will never reproduce exactly the possible repository situations such as the coupling of radioactivity, pressure and temperature. Therefore, in the best case, analogues can only contribute to safety cases as a line of evidence. They cannot be relied upon as the sole line of evidence. For instance, they can provide confirmation, with some associated uncertainty, of particular aspects in broader modelling or can aid in identifying the presence of specific phenomena such as chemical reactions, etc.

Overall, it is recognised among the technical community that analogues cannot be used as a sole and unique demonstration, but need to stand as one of multiple lines of evidence. As the degree of similarity with possible repository situations diminishes,

examples become analogies or anecdotes and their arguments are more useful in terms of supporting generic and conceptual feasibility of geological disposal.

## Analogues for public confidence?

Although analogues are often described as important confidence-building arguments for the public, most FSC members were not so affirmative in their assessment of this claim. Some typical quotes from the written input by FSC members in co-ordination with IGSC (safety case specialists) colleagues demonstrated scepticism:

- It is stated that "for technical specialists, analogues <u>are used</u>..." but the statement is not so affirmative related to the public: "towards the general public analogues <u>can be used</u>...".
- "We proactively use materials found in Japan to provide our readers a sense of familiarity. However, since their effectiveness measurement hasn't been carried out, we haven't figured out how much they've been helpful for confidence building."
- On one hand, "their main role for technical specialists is seen in their support...", but
  on the other hand, "their major role for the general public is probably to raise interest
  and increase understanding of processes by illustration and showing familiarity to
  many people."

It is this author's experience that analogues never played a role in the many peer reviews of national safety cases that the NEA has organised so far.

Nevertheless, most FSC members had the feeling that analogies and anecdotes could help the public to grasp time scale and understand the basic rationale and principles of geological disposal. They represent concrete examples of important functions such as long-term confinement properties to illustrate potential repository situations that might otherwise be seen as "pure theory".

## National vs. natural analogues

Some FSC members reported that the use of national analogues (i.e. drawn from situations within the host country) may be more effective for the general public than extra-national ones, notably when concerning anthropogenic analogues. Given that analogues are concrete representations of possible repository situations, it seems plausible that the public may be more likely to find a national analogue persuasive and comprehensible, since it is geographically and culturally closer to their own experience and likely to be very concrete, especially in the case of anthropogenic analogues. The familiarity with the case facilitates understanding and may itself provide some reassurance to the public. This finding was also reported by others at the present workshop on natural analogues.

## Analogues as a function of programme development

The FSC topical session appeared to indicate that the use of analogues as confidence-building arguments for the general public varies with time and with the stage of programme development and therefore depends on the decision-making process and its phases. As the project focuses on progressively more specific sites and more defined concepts, an increasingly closer relationship with the repository situations, for instance in terms of geological formations, engineered materials, etc., becomes possible. Later, as the project is accepted in its principle by local public stakeholders, it seems that the need for analogues, analogies and anecdotes for stakeholder confidence dwindles. This finding was confirmed at the present workshop on natural analogues. At the time, it was based only on observations in Finland and France, in which geological repository projects were quite ahead of those in other countries but still in relatively early phases compared to the 100 or more years in which they are meant to be an active presence in the host community.

#### Vision for the future

Possible developments that could be suggested included:

- Continue efforts to build analogues databases such as Nanet, notably for regulators and assessors, but as well in relation to the two following points:
  - Use such database matrices to integrate the so-called "negative" analogues and investigate thoroughly the cause of perceived discrepancies to transform the case into an added-value ("positive") analogue.
  - Translate the scientific and technical analogues, as inventoried in various databases, into convincing arguments for the public at large, with specific consideration for national analogues.
- · Measure the effectiveness of analogues and related arguments in terms of public confidence building, in order to improve the tools and techniques of presentation.
- Looking beyond only analogues, investigate the evolution over time (i.e. through the development of a repository) of the type of confidence-building arguments needed for the general public:
  - A first step would be to study whether any evolution has been observed with existing low- and intermediate-level, short-lived radioactive waste repository projects and, by extension, with other controversial industrial projects.

Analogues have been typically depicted to describe phenomena over time scales beyond hundreds of years. The application of contemporary analogues as input to the design and choice of materials with a view to suggest safe and reversible operation deserves attention. Indeed, the relationship between repository situations over a few centuries and contemporary analogues, such as tunnels or mines, is very close. Given the relatively early phase of most disposal projects, these analogues might be useful for discussing the concept of reversibility with wider audiences as well as with technical stakeholders. For instance:

- · The regulator may find this to be compelling evidence that reversibility is not achieved at the expense of safety.
- The public may be interested in demonstrations of reversibility in engineered systems, which could be provided through analogues.

Finally, and still in relation to the project phases, it was reported from Finland and France that current public issues in these countries tend to be socioeconomic ones. The topic of socioeconomic analogues (probably very contemporary ones involving similar projects in terms of investment, running costs, construction, operating life, footprint, etc.), which was not in this topical session's remit, could be a future theme of interest for the FSC and others.

## Reference

Organisation for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) (2008), "Link Between Research, Development and Demonstration (RD&D) and Stakeholder Confidence: Use of Analogues for Confidence Building", Proceedings of a Topical Session held during the NEA Forum on Stakeholder Confidence, NEA/RWM/FSC(2008)3, OECD/NEA, Paris. Also available on-line.

## **Session II**

## **Overview of National Programmes**

## NEA/RWM/R(2013)10

## The US government approach to management and geologic disposal of radioactive waste in the USA - 2013

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The US government released its "Strategy for the Management and Disposal of Used Nuclear Fuel and High-Level Radioactive Waste" (the Strategy) in January 2013 (USDOE, 2013). The Strategy is a policy statement that acknowledges that the previous federal government approach to siting major nuclear waste management facilities is not workable. Experience has shown that this previous top-down approach typically does not work if opposition is strong at high levels of local and regional/state government. The newly published strategy presents a way to create a more acceptable and sustainable programme to create an integrated system for transporting, storing and disposing of the higher-level wastes generated in both the civilian and the government sectors.

## Schedules for creating the components of the new US waste management strategy

In response to the recommendations from the final report and recommendations of the "Blue Ribbon Commission on America's Nuclear Future" (BRC) (BRC, 2012) the new strategy proposes a phased, adaptive and consent-based approach to siting and implementing a radioactive waste management and disposal system consisting of a pilot interim storage facility, a full-scale interim storage facility and a geologic repository.

The strategy proposes that over the next decade the following will be accomplished:

- site, design, license, construct and begin operating the pilot interim storage facility by 2021 with an initial focus on accepting used nuclear fuel from shutdown reactor sites:
- advance toward siting and licensing a larger interim storage facility, to be available by 2025;
- make demonstrable progress on the siting and characterisation of repository sites to facilitate the availability of a geologic repository by 2048.

## **Strategy elements**

As may be seen in Figure 1, taken from the Strategy, siting, design and governance/funding are its key elements. Each of the descriptive phrases in this figure reflects recommendations made in the final report from the BRC.

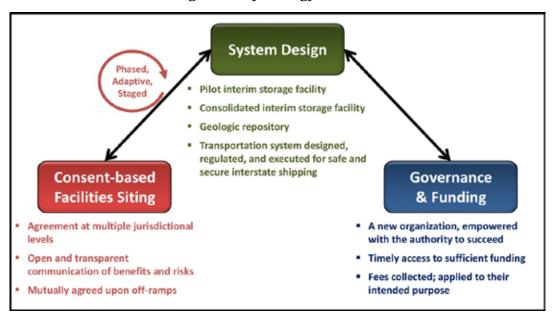


Figure 1: Key strategy elements

Source: US DOE, 2013.

## **Prospects for the future**

Perhaps in large part due to the positive manner in which the relationship between the host community for the Waste Isolation Pilot Plant (WIPP), the working deep geologic disposal facility in New Mexico, was described in the BRC report (2012), several locations in the United States, at several political levels, have indicated an interest in discussing how they might host a waste management facility as well. They have done this through written communications to the Department of Energy. These locations must be kept confidential for the present time.

The locations wishing to be considered for a deep geologic repository have proposed that sites in volcanic rock and sedimentary rock be considered. The majority of the proposed sedimentary rock sites that have been proposed for discussion and perhaps further consideration are in rock salt.

## Rock salt as a geologic repository medium

Since the currently operating US deep geologic repository is in rock salt (WIPP), it is a medium that is well studied and understood. However, because the current US repository is disposing of long-lived transuranic waste with no appreciable heat loading, some testing has been proposed to confirm the suitability of salt as a host media for the disposal of heat-bearing wastes. The proposal is in two phases; the first phase would be a field test that addresses the lesser heat output of defence and research reactor related (US government generated and owned) wastes (US DOE, 2012).

The second phase (US DOE, 2011) would focus on the effects from hotter (largely civilian reactor sourced) waste streams. Although this two-phased testing programme would be conducted at the WIPP, its results would be directly applicable to any proposed bedded salt site, and can also be useful for judging heat effects in any proposed domed salt sites. In other words, conducting the tests in one location does not make the tests site-specific. Since rock salt deposits are the primary (bedded) or physically reworked

(domed) remnants of seawater evaporation, many tend to have much in common physically and chemically, regardless of their location in the US or even the world.

The first phase of testing proposed is being defined, and will serve as a test bed for the design of the second phase. Therefore, only the conceptual layout for the first phase of testing will be illustrated here. This first phase focuses on the heat output expected from government generated and owned wastes currently being managed under the Department of Energy's Environmental Management (EM) programme. Figure 2(a) illustrates the layout for the testing. The concept for testing the lower heat output portion of the government owned wastes is illustrated in upper part of Figure 2(b), and the concept for testing the higher heat output portion of that same set of radioactive wastes is shown in the lower part of the same figure. The differences between both concepts lie in the spacing between the heater containers to control the temperature in the host rock. Instrumenting these tests to allow maximum data collection on processes of interest is a current challenge.

Figure 2(a): Conceptual layout for the government owned and managed waste heater tests showing two test drifts with different spacing between the heater containers

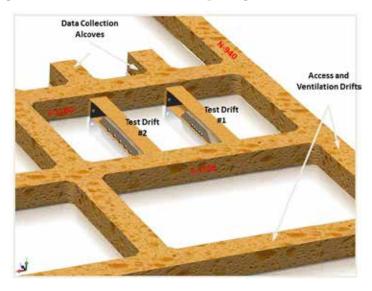
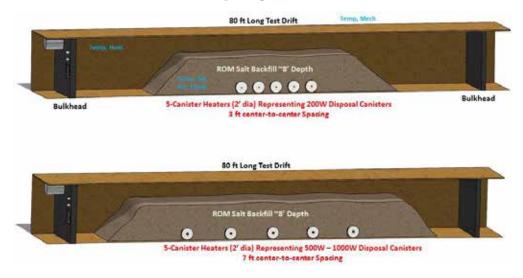


Figure 2b: Conceptual layout for Test Drifts 1 and 2 showing the two test drifts with different spacing between the heater containers



## Other geologic repository media under consideration in the United States

The Strategy (US DOE, 2013) indicates that the US focus for a future deep geologic repository will be on several types of host rock. Current work in the Department is looking at the potential for using crystalline intrusive or metamorphic rocks, and at sedimentary rocks like bedded or domed salt, shale or clay formations. The Strategy indicates that an effort will be made to co-ordinate with and learn from the repository programmes of other nations that have significant experience with these various geologic media.

## The current administration policy on the management of radioactive wastes reflects experience

The Strategy strongly suggests that the days of making a high-level decision about siting a nuclear waste facility are behind us (US DOE, 2013). Experience with a Nuclear Waste Negotiator in the 1990s and with siting the proposed Yucca Mountain repository during the last three decades indicates that in the US as elsewhere, smaller communities, counties or native American territories may show interest, but if at the state level there is strong opposition, there is no way to make progress in siting a facility. In the future, all levels of government with a say in the matter need to be consulted and co-ordinated with in order to achieve acceptance of a facility. The success of the Waste Isolation Pilot Plant Repository is hailed in the BRC report several times as a model for state to county to community involvement – a co-ordination leading to a successful outcome. In that process the federal government had to make adjustments to what it desired to do, the state imposed some limitations of waste volumes and types, but that is the whole idea behind a serious effort at consultation. In the end there was a binding written agreement between the state and the federal government that allowed the repository to open, and it has now been operating since 1999.

## A fundamental problem with siting nuclear facilities

The opposition to the siting of nuclear facilities is a world-wide phenomenon. Fear of radiation and radioactivity is the basis for this opposition, augmented by very rare nuclear disasters. This fear exists in the public, in politicians, scientists and regulators. Fear of radiation, and the precautionary way that some radiation contamination incidents have consequently been managed, in some instances has been more detrimental to the affected public's well-being than the radiation itself.

Perhaps a fertile area for future co-operation between nations is to create effective ways to bring understanding into schools and other appropriate public educational forums on radiation, its nature, sources and effects. Part of this curriculum should explain the precautionary nature of national and international regulations that assume effects from low-doses of radiation that have been predicted, but have never been observed in normally healthy whole organisms.

## References

- Blue Ribbon Commission on America's Nuclear Future (BRC) (2012), Report to the Secretary of Energy, accessed 9 April 2013,
  - http://cybercemetery.unt.edu/archive/brc/20120620215759/http://brc.gov/index.php?q=meetings/765.
- United States Department of Energy (US DOE) (2013), Strategy for the Management and Disposal of Used Nuclear Fuel and High-Level Radioactive Waste, Washington, DC, accessed 8 April 2013,
  - http://energy.gov/sites/prod/files/Strategy%20 for%20 the%20 Management%20 and%20 Disposal%20 of%20 Used%20 Nuclear%20 Fuel%20 and%20 High%20 Level%20 Radioactive%20 Waste.pdf.
- US DOE (2012), A Conceptual Plan for Salt Defense Disposal Investigations for the Disposal of DOE-EM Managed Wastes, DOE/CBFO-12-3485 Rev. 0, Carlsbad Field Office, Carlsbad, New Mexico, accessed 9 April 2013, www.epa.gov/radiation/docs/wipp/sdi/doe\_sdi\_62012-2.pdf.
- US DOE (2011), A Management Proposal for Salt Disposal Investigations with a Field Scale Heater Test at WIPP, DOE/CBFO-11-3470 Rev. 0, Carlsbad Field Office, Carlsbad, New Mexico, accessed 9 April 2013,
  - www.wipp.energy.gov/library/EPA/DOE-CBFO-11-3470\_Rev\_0.pdf.

## NEA/RWM/R(2013)10

## The current status of the geological disposal of radioactive waste in Poland

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There are plans to build the first Polish nuclear power plant by 2025. Finding a site for low- and intermediate-level waste (LLW, ILW) is currently a top priority in Poland. Candidate sites for a deep geological repository to host high-level waste (HLW) and spent nuclear fuel (SNF) from the Polish NPPs selected during the strategic governmental programme in 1999 are still valid. They include Triassic shale and two Zechstein salt domes. No ranking of the candidate sites has been done at this stage of site selection because of the lack of necessary data. Therefore, detailed and costly geological and geophysical investigations including drilling are required to further evaluate the proposed candidate sites. Salt domes or bedded salt may be the best option for the Polish geological repository. The Kłodawa salt dome seems to be suitable for building the underground research laboratory (URL) to study problems relevant to disposal of radioactive waste in Polish salt domes and salt structures in general. The unique feature of Polish salt domes is the occurrence of large volumes of salt mudstones that can be used as backfill material due their favourable mechanical and sorptive properties.

## Introduction

Poland does not yet have a nuclear power plant (NPP). However, there are plans to build one by 2020 or 2023. The Polish government seems to be strongly committed to pursue the Nuclear Power Program first issued in 2010 and currently updated. Actions have been undertaken accordingly, including site selection for NPP. According to the Nuclear Power Program, nuclear fuel will account for almost 16% of all types of fuel used to generate electricity in Poland in 2030.

Currently, the site selection process for NPP is in progress. Candidate sites have been selected based on general criteria recommended by the International Atomic Energy Agency (IAEA). All major candidate sites are situated either in northern Poland or in north-central Poland (Figure 1). The site at Żarnowiec (pronounced Zharnovets) is of particular interest. Żarnowiec was selected as the site for nuclear power plant in the 80s and the construction of the power plant began in the latter part of that decade. Some 30% of the project had been completed before the construction was terminated in 1990 amidst social protests and concerns related to nuclear safety issues after the Chernobyl disaster in 1986. However, the site seems to be suitable for building the nuclear power plant and is the "first pick" of the Polish Energy Group – a company responsible for siting. Issues related to radioactive waste management are outlined in the National Nuclear Power Program. Two actions in the Program are devoted to the final stage of the nuclear fuel cycle, i.e. radioactive waste and SNF management, one aimed at finding a site for a repository for LLW and ILW by 2020, and the other aimed at preparing a national plan for HLW and SNF management.

The Ministry of Economy is responsible for radioactive waste management in Poland, whereas the National Atomic Energy Agency (PAA), supervised by the Ministry for the

Environment, is responsible for nuclear and radiation protection. Under Polish law, the PAA has the right to license all activities related to nuclear energy and nuclear materials in Poland. Radioactive waste (LLW and ILW) generated by industry, research laboratories (including two research nuclear reactors, one of which was decommissioned between 1997 and 2002), hospitals and other users are handled by the Radioactive Waste Neutralisation Plant (ZUOP) near Warsaw.

The LLW waste is disposed of in the only Polish repository (National Radioactive Waste Repository, KSOP) in Różan some 30 km NE of Warsaw. The repository, established in 1961, is hosted by an old military fort (built in 1910) and will be filled in with solid LLW and ILW by 2020 or 2022 (Cholerzyński, 2012). Therefore, finding a new site for a subsurface repository for LLW and ILW is a top priority for the Polish authorities responsible for waste management.

As for the HLW and SNF from a planned NPP, the National Plan is now being prepared by a working group under the supervision of the Ministry of Economy. A deep underground (geological) repository is of no immediate concern in the Polish nuclear power programme for two main reasons: i) SNF from two research reactors is safely stored on-site at the Radioactive Waste Neutralisation Plant (ZUOP). Highly enriched SNF from the interim storage facility was transferred to Russia in 2010 under the agreement signed in 2009 by Poland, the United States and Russia. SNF from another interim storage facility will be shipped to Russia within next few years (Włodarski, 2012). ii) SNF from the first Polish NPP will have to be disposed of around 2050. The relatively long time span between the beginning of the NPP operation and the necessity of disposing of HLW and/or SNF makes this issue less one of timing from the perspective of the authorities. Moreover, a preliminary search for the geologically most suitable sites for a deep underground repository had already been done in Poland 13 years ago.

## Preliminary results of site selection for a deep geological repository in Poland

A Strategic Governmental Program for radioactive waste and spent nuclear fuel management in Poland was conducted from 1997 to 1999 aiming at site selection for both a near surface and a deep geological repository (Janeczek and Wlodarski, 2001). Results of the Programme are still valid. During the first stage of the siting procedure, 44 sites in various rock formations were selected for further consideration. After a more detailed screening based on archive geological data supplemented by new geophysical research, environmental analysis, and socio-economic analysis, the seven most promising sites were selected: three in granitic rocks within the basement of the Eastern European Platform in NE Poland, two in salt domes and one in the clay formation of the Fore-Sudetes Monocline (Figure 1).

Knowledge of the geology of the deep geological formations in NE Poland is not detailed enough for the evaluation of their suitability for hosting a geological repository. Therefore, while not entirely rejected, the granitic rocks in the basement of NE Poland are considered to be of secondary interest in the site selection process.

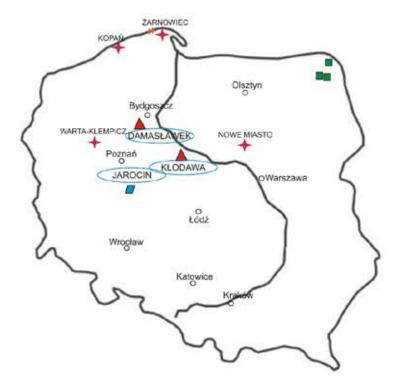
Based on general criteria for radioactive waste disposal in shale, a candidate site was selected within the Triassic clay formation known as the Upper Gypsum Beds (UGB) near Jarocin (Figure 1). The UGB belong to a large geological unit called the Fore-Sudetic Monocline and consist of almost horizontal (dipping at 2 degrees) layers of shale and clay stone composed predominantly of illite and Mg-chlorite. They occur at an average depth of 535 m. Their thickness is over 200 m and they are overlain by a 300 m thick sequence of practically impermeable shale and mudstone, which isolate UGB from the uppermost water-saturated Tertiary and Quaternary sediments (Slizowski, 1999a). The UGB are underlain by permeable sandstones. Beneath them there is impermeable shale. Therefore, the overall hydrogeological conditions are favourable for disposal of radioactive waste.

Figure 1: Location of candidate sites for NPP and geological repository in Poland

Candidate sites for NPP: preferred sites – large stars; secondary candidate sites – small stars.

Candidate sites for GR: salt domes – triangles; shale – diamond; granitic rocks – squares.

Encircled sites are discussed in the text.



Source: Compiled from the Polish Nuclear Energy Programme (2010) (candidate NPP sites) and from Janeczek and Wlodarski (2001) (candidate geological repository sites).

Based on information from boreholes it was possible to outline a monolithic block within UGB with a diameter of some 2 km that may host disposal rooms of the repository. Because the type of nuclear fuel cycle to be implemented in the Polish NPP has not yet been decided upon, optional emplacement methods have been proposed separately for vitrified HLW (the in-floor borehole emplacement of waste packages) and for unprocessed SNF (the in-room emplacement of SNF packages) (Slizowski, 1999b). In case of disposing of vitrified HLW, the boreholes have to be inclined due to the limited thickness of the clay strata.

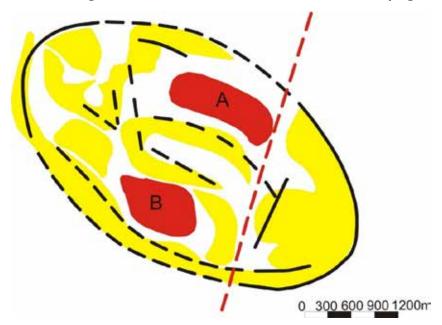
The Zechstein salt belt that hosts German repositories at Gorleben and Morsleben extends to Central Poland. There are numerous salt domes beneath the Polish Lowland. Two of them have been considered as candidate sites for geological repositories based on IAEA basic criteria for site selection in salt domes (Figure 1). Special attention was paid to the salt dome near the village of Damasławek SW of the city of Bydgoszcz.

The Damasławek salt dome protrudes through the Mesozoic sedimentary rocks into the Miocene formation from the depth of ca. 2.5 km (Krzywiec, 1999). Interpretation of geophysical data suggests that the salt dome formed in the Upper Cretaceous. Before the Cenomanian and Turonian, salt intruded overlying strata and reached the surface of the bottom of the sedimentary basin. As a result the uppermost part of the salt dome was partially dissolved and the old upper cap rock formed. In the Paleogene, the young lower cap rock formed. In the Miocene the salt dome subsidised some 100 m and then was uplifted again in the late Tertiary and in the Quaternary. The rate of the recent salt dome

uplift has been estimated at 40 m per one hundred thousand years (Krzywiec, 1999). The roof of the cap rock occurs at a depth of between 180 m and 260 m below ground level. The 200 to 240 m thick cap rock is heterogeneous and consists of anhydrite, gypsum and clay. The uppermost part of the cap rock is heavily fractured and faulted. Fractures are water conducive and faults can be rejuvenated. The salt table occurs at a depth of 460-480 m. Based on geophysical data and on archive geological data from drill holes the most promising location has been suggested to be at a depth of 800 m, within homogeneous and coarse-grained rock salt away from major fractures and faults in overlying cap rock (Krzywiec, 1999). The faults do not propagate into the coarse-grained salt. The selected area is located in a region with the least vertical movements in sedimentary rocks above the salt dome (Figure 2). The overlying cap rock is completely dry and undisturbed.

Figure 2: Sketch map of the Damaslawek salt dome at the depth of 800 m b.g.l.

A and B – areas within unfractured, homogeneous salt rock most suitable for hosting geological repository (A is a preferred location); solid lines – youngest faults propagating through Miocene rocks into the rock salt; broken lines – faults in cap rock ending in overlying Miocene rocks; shaded areas – areas with the largest difference in thickness of Miocene strata overlying the salt dome.



Source: Krzywiec, 1999.

The size and geometry of the selected area within the salt dome are suitable for the planned volume of radioactive waste to be disposed of in disposal rooms. The selected part of the rock salt is situated far away enough from the dislocation boundaries of the salt dome. The conceptual model of the repository in the salt dome was proposed with the disposal rooms at a depth of some 800 m below ground level. As with the case of a repository in clay formation, the optional methods for emplacement of waste packages have been proposed separately for vitrified HLW and for unprocessed SNF (Ślizowski, 1999b). Unconfirmed reports suggest that the Damasławek salt dome may be used for underground gas storage. If this is the case, that salt dome will be withdrawn from the list of potential sites for the geological repository.

Another candidate site is located in the Kłodawa salt structure near an active underground salt mine (Figure 1). The Kłodawa salt dome is a part of a 63 km long salt structure (Figure 3). Due to mining activities this is the best recognised salt dome in

Poland (Ślizowski, 1999b). Natural hazards in the proposed site for a geological repository will most likely be the same as those for the Kłodawa salt mine (methane ejections in mine workings, and water-saturated Tertiary sands and Quaternary sediments above the salt dome pose a major threat to the mine). Hydrogeological conditions in the Kłodawa salt mine have not caused significant problems to the safety of mining for 60 years. The Kłodawa salt dome is composed of folded rock salt intercalated with thin layers of potassium salts, anhydrite and a thick sequence of salt-mudstone (500-600 m). The roof of cap rock is at a depth of 200-300 m below the ground surface. The overall thickness of the gypsum-anhydrite-clay cap rock is between 100-150 m. The repository can be located some 2 km away from the tunnels and chambers of the salt mine at a depth between 600-1 000 m. A two-level repository can be built in the salt dome because of the large thickness of both salt and salt mudstones (Ślizowski, 1999b).

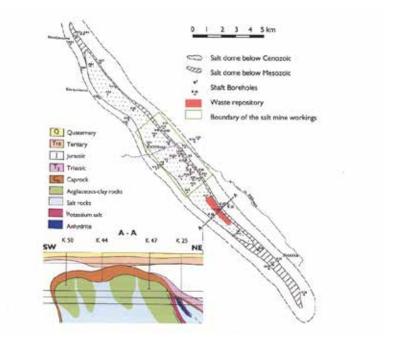


Figure 3: Geological map and cross-section of the Kłodawa salt dome

Source: Ślizowski, 1999b.

A project for an URL in the Kłodawa salt dome was proposed (Ślizowski, 1999c). One of the rationales for this laboratory is to enable in situ examinations of mechanical and thermo-mechanical properties of salt mudstones. The unique feature of Polish salt domes is the occurrence of rocks that consist of salt, anhydrite, quartz and clay minerals in various proportions. These rocks have collectively been called "zuber" by miners and by Polish salt geologists. They have been extensively investigated as potential host rocks for a geological repository (Ślizowski, 2005).

There are two types of salt mudstone that differ in colour (red and dark brown) and in contents of anhydrite and quartz, as well as of some minor minerals (Table 1). The ratio of rock salt to non-soluble minerals varies considerably from sample to sample within short distances in the salt dome. Both varieties of "zuber" rocks have the same concentrations of clay minerals, which also vary considerably. Clay minerals are equal amounts of Mg-rich chlorites and illite/muscovite (Bzowska and Janeczek, 2005). The sorption capacity of salt mudstone rocks increases with increasing amount of clay minerals as evidenced by experiments in which <sup>90</sup>Sr and <sup>152,154</sup>Eu were sorbed by halite and by numerous samples of "zuber" rocks with different proportions of clay minerals (Gilewicz-Wolter, et al., 2005).

Table 1: Mineral composition (vol.%) of salt mudstone ("zuber") from the Kłodawa salt dome

Brown '	ʻzuber"	Red "zuber"		
Halite	8-76	Halite	20 88	
Anhydrite	Anhydrite Up to 75		Up to 50	
Clay minerals	Up to 25	Clay minerals	Up to 25	
Magnesite	Up to 10	Magnesite	0-25	
Dolomite	Not detected	Dolomite	3-10	
Hematite	Not detected	Hematite	1-4	

Source: Bzowska and Janeczek, 2005.

In addition to true sorption, ion exchange with  $Ca^{2+}$  and precipitation of  $SrSO_4$  are mechanisms for  $^{90}Sr$  immobilisation by "zuber". The "zuber" rocks combine the sorptive properties of clay minerals with mechanical properties of salt and, therefore, have been suggested as suitable host rock for construction of the geological repository (Ślizowski, et al., 2003). However, their inhomogeneity poses some serious doubts about their usefulness as a host rock for a geological repository. Moreover, geomechanical properties of salt mudstones, particularly long testing endurance under elevated temperature lower than for rock salt and low values of the deformation stress at the destroying point may cause serious problems during construction of the deep repository (Ślizowski, et al., 2005). On the other hand, "zuber" rocks can certainly be used as a backfill material in a geological repository.

Currently, disposal of radioactive waste in thick bedded Zechstein salt in northern Poland is under consideration as an alternative to the salt domes described here. Relatively simple geology, lack of significant tectonic deformations, favourable hydrogeological conditions, thickness in the range of 150-225 m and location at a depth of ca. 650 m (Ślizowski and Lankoff, 2009) make bedded salt a promising option.

### **Concluding remarks**

Finding a site for LLW and ILW is currently a top priority in Poland. Candidate sites for deep geological repository to host HLW and SNF from Polish NPPs selected during the Strategic Governmental Programme in 1999 are still valid. No ranking of the candidate sites has been done at this stage of site selection because of the lack of necessary data. Therefore, detailed and costly geological and geophysical investigations including drilling are required to further evaluate the proposed candidate sites. Salt domes or bedded salt in northern Poland may be the best option for the Polish waste management programme. The Kłodawa salt dome seems to be well suited for building the URL to study problems relevant to the disposal of radioactive waste in Polish salt domes and salt structures in general. Finally, the lack of acceptance for the preliminary selected sites in 1999 from the affected (local) population is noticeable. Therefore, gaining societal acceptance for the geological disposal of radioactive waste in Poland is equally important to technical issues.

## References

- Bzowska, G. and J. Janeczek (2005), "Mineralogical Investigation of Minerals Other than Salt; In Particular, Clay Minerals", in Laboratory Studies of Zubers (Salt Mudstones) for Evaluation of Suitability for Disposal of Radioactive Waste in Polish Salt Domes, K. Slizowski (Ed.), IGSMiE PAN Cracow, 37-43 (in Polish).
- Cholerzyński, A. (2012), "Closure of National Radioactive Waste Repository in Różan (KSOP-Różan) Preparation of Long-Live Waste for Storage in Another Repository", Nuclear Safety and Radiological Protection, No. 3, Polish Atomic Energy Agency, 7-13 (in Polish).
- Gilewicz-Wolter, J., et al. (2005), "Immobilization of Selected Radionuclides in Salt Mudstones" (in Polish), in Laboratory Studies of Zubers (Salt Mudstones) for Evaluation of Suitability for Disposal of Radioactive Waste in Polish Salt Domes, K. Slizowski (Ed.), IGSMiE PAN Cracow, 49-58 (in Polish).
- Janeczek, J. and J. Włodarski (2001), "The Polish Concept of Radioactive Waste Disposal", in Geological Challenges in Radioactive Waste Isolation, Third Worldwide Review, P.A. Witherspoon and G.S. Bodvarsson (Eds.), Ernest Orlando Lawrence Berkeley National Laboratory, University of California Berkeley (LBNL-49767), 205-208.
- Krzywiec, P. (Ed.) (1999), Internal Structure of the Gypsum-Anhydrite Cap Rock in the Damasławek Salt Dome Investigated by Shallow Seismic Profiling, Unpublished Technical Report, Polish Geological Institute, Warsaw (in Polish).
- Polish Nuclear Power Programme (2010), Ministry of Economy, Warsaw (in Polish).
- Polish Atomic Energy Agency (PAEA) (2000), Strategic Governmental Programme: Radioactive Waste and Spent Nuclear Fuel Management in Poland, Unpublished Final Report, Polish Atomic Energy Agency, Warsaw.
- Slizowski, J., W. Kasprzyk and L. Lankof (2005), "Rheological Properties of Salt-Mudstones", in Laboratory Studies of Zubers (Salt Mudstones) for Evaluation of Suitability for Disposal of Radioactive Waste in Polish Salt Domes, K. Slizowski (Ed.), IGSMiE PAN Cracow, 73-82.
- Slizowski, K. (Ed.) (1999a), Evaluation of the Possibility to Dispose of Radioactive Waste in a Deep Underground Repository in Clay Stones of the Fore-Sudetic Monocline in the Jarocin-Pogorzela Area, Unpublished Technical Report, Institute of Mineral Resources and Energy Management, Polish Academy of Sciences, Cracow (in Polish).
- Slizowski, K. (Ed.) (1999b), The Conceptual Model of the Polish Deep Underground Radioactive Waste Repository in (a) Rock Salt, (b) Shale, Unpublished Technical Report, Institute of Mineral Resources and Energy Management, Polish Academy of Sciences, Cracow (in Polish).
- Slizowski, K. (Ed.) (1999c), Conceptual Model for Underground Research Laboratory in the Kłodawa Salt Dome, Unpublished Technical Report, Institute of Mineral Resources and Energy Management, Polish Academy of Sciences, Cracow (in Polish).
- Slizowski, K., J. Janeczek and K. Przewłocki (2003), "Suitability of Salt Mudstone as a Host Rock in Salt Domes for Radioactive-Waste Storage", Applied Energy, 75, 119-128.

## NEA/RWM/R(2013)10

- Slizowski, K. (Ed.) (2005), "Laboratory Studies of Zuber (Salt Mudstones) for Evaluation of Suitability for Disposal of Radioactive Waste in Polish Salt Domes", IGSMiE PAN Cracow (in Polish).
- Slizowski, K. and L. Lankoff (2009), "Geological Conditions of High Level Radioactive Waste Storage in Polish Salt Deposits", Prz. Geol., 57, 829-838 (in Polish).
- Włodarski, J. (2012), "Information of the Polish Atomic Energy Agency about Nuclear Safety and Radiological Protection in Poland in 2011", Nuclear Safety and Radiological Protection, No. 2, Polish Atomic Energy Agency, 3-54.

# Status of R&D programme on geological disposal of radioactive waste in the Netherlands

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The Netherlands has a policy of interim storage for a period of at least 100 years. During this period, disposal is to be prepared for, socially, technically and economically, such that it can be implemented after the interim storage. After a period of almost 10 years without a national research programme, the challenge was to restart the research on geological disposal. This paper describes the Dutch (research) policy, the previous research programmes, the restart of the national programme in 2011, and the current status of the third national programme.

### Introduction

The radioactive waste policy in the Netherlands is that all kinds and categories of radioactive waste are managed by one central waste management organisation (COVRA¹) and stored for at least 100 years at one site, above ground in engineered structures. This allows retrieval at all times. This step is to be followed by geological disposal for all categories (low-, intermediate- and high-level waste) in one single repository. During the period of interim storage, disposal is to be prepared for socially, technically and economically, such that it can be implemented afterwards.

Implementing and operating a small repository is costly, in particular for countries with small nuclear power programs such as the Netherlands. The economy-of-scale will force them either to implement long-term storage and wait for decades, and/or to share a repository with others. With only 525 MWe installed nuclear capacity in the Netherlands, two research reactors, an enrichment facility and about 200 producers of institutional waste, there is no need for disposal in the short term. The volume of all categories of radioactive waste generated over 30 years is only a few thousand m³: 60 m³ of HLW, 10 000 m³ of LILW and another 10 000 m³ of NORM waste. The resulting disposal costs per m³ are very high as long as the accumulated amount of waste is small.

The additional costs of prolonged interim storage are relatively small and the small volume of waste can easily be kept under control in above-ground structures. This "interim" storage provides time to let the volume of waste accumulate and to let the amount of money needed for disposal to grow in a capital growth fund. In 100 years' time, growth by about a factor of 10 can be obtained with a real interest rate of 2.3%. Moreover, an international or regional solution may become available during the next 100 years.

Long-term storage is currently in full operation and necessary provisions for the next step have been taken as well. The capital growth fund to finance final disposal exists, waste generators pay for this and there is a clear choice for the ownership of the waste:

<sup>1.</sup> The Centrale Organisatie Voor Radioactief Afval (CORVA) is the central organisation in the Netherlands for collection, treatment, storage and eventual disposal of radioactive wastes.

all liabilities are transferred to the waste management organisation COVRA. The challenge was, however, to restart the research on geological disposal after a period of almost 10 years without a national research programme. This paper summarises the Dutch (research) policy, looks back on the previous programme, and describes the restart of the national programme in 2011 and its current status.

#### Discussion

## Research policy

The long-term waste management strategy in the Netherlands is based on the main components of the Dutch policy regarding radioactive waste management. This policy, first recorded in governmental documents in respectively 1984 (VROM, 1984) and 1993 (Dutch Government, 1993) will be briefly outlined below. In addition to the issues of minimisation and reduction of (the amount of) radioactive wastes the policy in the Netherlands has two main components:

- · interim storage of all kinds of radioactive wastes at a centralised site of COVRA for at least 100 years;
- · ongoing research on retrievable disposal in deep geological formations;

The objectives of the research were laid down in a policy paper on radioactive waste that was discussed and accepted by Parliament in 1984: "Therefore a site must be found in the Netherlands where storage of all categories of radioactive waste can take place. During the storage period further considerations can be given to final disposal, international developments can be followed and even an international facility could be used."

In the 80s an extensive research programme was started with the goal to build an underground test facility in the Netherlands. Feasibility studies and safety studies concluded that building a disposal facility is technically feasible and that underground disposal of the waste is safe. The safety studies involved desktop studies as well as experimental studies in the Asse research facility. The safety studies involved two iterations: the first study (VEOS) showed on a deterministic basis that the dose rates in the biosphere would always remain very low for various scenarios. The second study (PROSA) followed and expanded on international practice in the area of probabilistic safety analysis as applied to geological disposal. In addition, it was attempted to use all international efforts in regarding the definition of FEPs and to develop the scenarios to be addressed probabilistically on a systematic basis.

Research on a retrievable disposal is an important prerequisite of the strategy, in line with the (fourth) National Environmental Policy Plan (NMP, VROM, 2001) in which the government established a strategy for sustainable development. The Dutch policy plan, which stated that eventually underground disposal is permissible if the waste remains retrievable over the long-term, was a consequence of a NMP initiative implemented in 1993.

Long-term retrievable disposal of radioactive waste is considered to be feasible in both available host environments in the Netherlands: rock salt formations and clay layers (CORA, 2001). The CORA³ Commission, which was appointed by the Dutch government to co-ordinate the R&D programme, recommended further investigations on societal and ethical issues regarding waste management, in addition to the ongoing technological research.

<sup>2.</sup> Retrievability means the deposition of radioactive waste in a way that it is reversible for the long-term by proven technology without re-mining.

<sup>3.</sup> Commission for Radioactive Waste Disposal; in Dutch: Commissie Opberging Radioactief Afval.

The responsible ministry in the Netherlands (VROM<sup>4</sup>, now economic affairs) reflected on these recommendations with a policy document that describes the government's perspective on radioactive waste management (VROM, 2001). This policy envisages expanding knowledge in an international framework on all aspects of (deep geological) disposal, including technological, societal and ethical issues, during the interim storage phase. Keeping all options open is also part of this strategy, which avoids any exclusion of potential waste management opportunities by taking (irreversible) decisions now.

The ongoing research on disposal in deep geological formations is line with the EC directive (2011/70/EURATOM) that requires a national programme for the safe and responsible management of spent fuel and radioactive waste. The national programmes should contain all research, development and demonstration activities necessary to implement the solutions for management of radioactive waste.

## Previous research on geological disposal

In the Netherlands, research on radioactive waste disposal started in the early 70s. It was pointed out that rock salt formations in the Netherlands could serve as host rock for a disposal facility. In 1993 the Dutch government issued a policy directive, which states that underground disposal of highly toxic waste (including high-level radioactive waste) is permitted provided it is retrievable for a long period of time. Therefore, the R&D programme was adjusted to focus on three options for retrievable storage or disposal: long-term above-ground storage (up to 300 years) or underground in either rock-salt formations or deep clay deposits (assumptions: a retrievable disposal in clay of rock salt at 500 m depth, with 50 m and 200 m of surrounding host rock, respectively). The CORA Commission (Commission on Radioactive Waste Disposal) was established to co-ordinate the R&D programme. In this programme, for each of three waste management options under consideration, the retrievability and safety aspects have been evaluated.

Two investigated host formations for deep geological disposal of radioactive were the rock salt formations mainly located in the northeast part of the country and (Boom) clay layers of which the studied depth can be found in the south near the Belgium border. The investigated disposal concepts contains short horizontal disposal cells drilled into the sidewalls of a gallery, each accommodating one HLW container. The annular space around the container is filled with suitable backfill material and the cell is sealed off. Retrieval requires a special machine to remove the seal and backfill and to withdraw the waste container from the disposal cell. Figure 1 shows an artist's impression of the reference design in rock salt.

The CORA research programme comprised 21 projects, with contributions from 20 research institutes both in the Netherlands and abroad. The programme was carried out during the period from 1996 to 2000 at a cost of about EUR 3.5 million. The CORA report is available in Dutch, with an executive summary in English.

For underground disposal, the studies resulted in the expectation that implementation of the condition for retrievability of the waste is feasible for both host rocks without compromising safety. Recommendations for future research include social-ethical aspects (criteria, stakeholders, stepwise process), more research into clay and international co-operation; the research should be implementer-driven. Despite the recommendations having been discussed and agreed upon by the parliament in 2001, and budget for follow-up research having been reserved, the follow-up programme was not started due to differing opinions between the Ministry of Economic Affairs and VROM about the financing of the programme.

67

<sup>4.</sup> Volkshuisvesting, Ruimtelijke Ordening en Milieubeheer (Housing, Spatial Planning and the Environment).

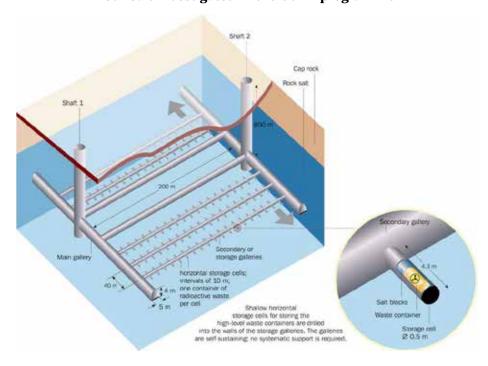


Figure 1: Retrievable disposal concept in rock salt investigated in the CORA programme

Source: CORA, 2001.

## **Restarting research (OPERA)**

From collaboration between NRG and COVRA on geological disposal research projects grew the idea to restart the Dutch national research programme on geological disposal. Based on the CORA recommendations, consultation with research institutes, and a workshop (ministries, universities, research institutes, nuclear organisations, ONDRAF/NIRAS and COVRA), COVRA and NRG together drafted an outline for a five-year research programme in 2007 that had broad support among stakeholders in the Netherlands. Nevertheless, it took a year and a half to get agreement on the financing. Finally in 2009, it was agreed to split the required EUR 10 million for the programme between industry (50%) and the government (50%).

When the plan received a green light, restarting the research after years without an active programme proved to be challenging. As the programme was to be implementer-driven, a dedicated management organisation within COVRA had to be set up. At the same time, the outline of the research programme had to be elaborated into a concrete project, including well-defined objectives and scope, five-year planning, a research plan and a first outline of a disposal concept. Due to these preparations, the programme did not actually get underway until 2011. The objectives and scope, organisation, planning and research plan, and the disposal concept are briefly described below.

## Objectives and scope

The starting points for defining the objectives and scope were: resolving outstanding issues from previous programmes, developing and preserving expertise and knowledge, and being prepared for site selection in case of any change in the current timetable, arising by way of future European directives, for example. Research activities are also required on the basis of Article 8 of the EC Directive on radioactive waste as a way to obtain, maintain and develop necessary expertise and skills for the management of radioactive waste.

The aim of OPERA, therefore, is detailing a first roadmap for the long-term research on geological disposal of radioactive waste in the Netherlands. This roadmap should be based initially on a re-evaluation of existing safety and feasibility studies conducted more than ten years ago, making use of present international and (wherever possible) national knowledge. In particular it was decided to build on the ONDRAF/NIRAS disposal concept and research on Boom clay. The Belgian programme has developed extensive knowledge of disposal of radioactive waste in Boom clay since 1974. The Belgian programme includes an underground research laboratory in Mol where experiments have been and still are performed to validate models.

The objectives are firstly to make a structured assessment of the long-term safety of a generic GDF in Zechstein rock salt and Boom clay formations in the Netherlands, and secondly to investigate the retrievability of waste from the facility and the technical feasibility of the proposed design. Societal and ethical processes that influence the acceptance and the required safety levels are considered to be an essential part of the framework for this assessment. To distinguish between the safety cases from advanced programmes (from e.g. Finland and Sweden), it was decided to employ the terms "initial" and "conditional" safety cases. "Initial" safety cases as they are intended to mark the start of the research development process and to iterate these as knowledge grows to new developed insights. "Conditional" safety cases since particular aspects including operational aspects have a smaller priority at this stage or are not considered at all and the outcome is based on certain conditions (plausible assumptions that must later be confirmed in a full safety case). For example, because the Netherlands has adopted the strategy of storage of radioactive waste for at least 100 years, it was decided not to consider site selection in OPERA.

A further aim of the programme is building and maintaining the knowledge and competences to run a geological disposal research programme and manage the interfaces between all steps of the radioactive waste management process from generation to disposal. Objectives are to reactivate research on geological disposal and to involve a broad group of (new) researchers in the field, as well as to provide access to previous research on geological disposal in the Netherlands, communicate transparently about the results and embed the (developed) knowledge in an academic curriculum. OPERA results and reports will be published at the COVRA website (ww.covra.nl).

## Organisation

In order to manage the programme, a new organisation had to be set up (Figure 2). A steering group had to be established first to monitor and steer the programme and control the programme finances; it consists of representatives of the government (Ministry of Economic Affairs) and the energy sector (the utilities EPZ, Delta and Essent). Its task was to approve the programme organisation. It was decided to clearly separate the tasks of carrying out the research from the task of managing the programme, and to keep the management cost as low as reasonably possible.

Within COVRA a small unit was established to manage the programme, co-ordinate the research projects and develop the safety cases. For the development of the safety cases the management is supported by the Safety Case Group. This group combines both international experts as well as national experts, who were involved in the previous national programmes to ensure inter-programme consistency. The management can also call for technical audits of research proposals and results by experts from abroad.

An Advisory Group was established to advise the management on the social relevance of the research, on communication and to propose new research projects. This group consists of stakeholders (representatives from drinking water companies, provinces) and academia (independent professors in ethics and risk communication, and geohydrology). The members of the group were appointed directly by the Minister of Economic Affairs.

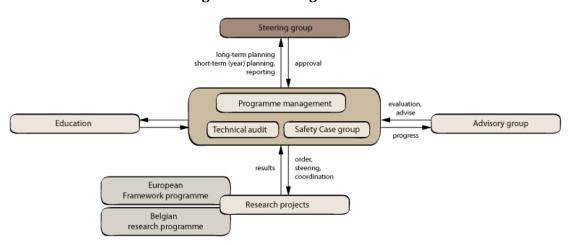


Figure 2: OPERA organisation

OPERA also collaborates with Delft University of Technology to integrate the programme outcomes in an academic curriculum and sponsors a chair on the Chemistry of Nuclear Fuel Cycle in the Master of Science Engineering. In this way, OPERA results and expertise are directly used to train new engineers. Finally, in order to involve a broad group of (new) researchers in the field, it was decided to ask for research projects through two public calls for proposals. This required, however, a detailed description of the research tasks required and an outline of the disposal concept investigated to make sure the different research projects are complementary and add up to make the safety case.

### Five-year planning and research plan

Five-year planning was first developed as a guideline for the execution of the research. The plan contained a clear frame for programming on a yearly basis, including criteria for selection of topics, the priorities and a logical order of research topics and a structure for setting up the calls for research projects. It also described an outline for communication about OPERA. Complementary to the five-year planning, a research plan was developed together with NRG.

The research plan elaborated the research programme description in the planning. It described the structural elements of the safety cases upon which the needs for R&D are based and the research necessary to develop two initial, conditional safety cases for national repository concepts for radioactive waste in rock salt and in Boom clay. In meetings with different Dutch research institutes [such as NRG, ECN and TNO and Dutch universities such as Delft University of Technology, Utrecht University and Wageningen University and the Energy Research Centre (ECN)] the research topics in over 40 complementary tasks with well-defined content and clear interfaces with other tasks, were grouped into seven work packages. This facilitated calling for proposals through public calls.

The research plan in the developing stage was discussed in the workshop with the Dutch research institutes and the Belgian agency for the management of radioactive wastes ONDRAF/NIRAS, and finally reviewed by an external reviewer (MCM Consulting).

## Disposal concept

In order to help the visualisation of the geological repository for the various research groups taking part in OPERA and to provide context for external communications, an outline disposal concept for Boom clay was developed in collaboration with NRG. The study of disposal concepts in salt will rely on concepts that were developed under the previous research programme CORA (CORA, 2001; Hijdra, 2000; Poley, 2000).

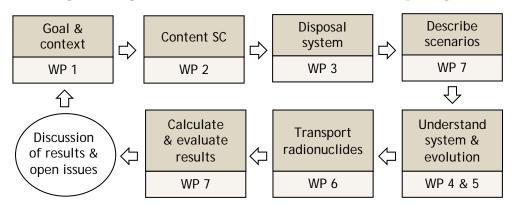


Figure 3: Organisation of the research into seven work packages

The OPERA supercontainer was based on the Belgian supercontainer concept, which consists of a carbon steel overpack, a concrete buffer and stainless steel envelope and can hold two HLW canisters or one SF canister (Humbeeck, et al., 2007). In OPERA a uniform supercontainer is used for the heat-generating HLW, spent fuel from research reactors as well as the non-heat-generating HLW. Figure 4 shows an artist's impression of the OPERA supercontainer for heat-generating HLW Alternatively, a supercontainer without the steel envelope will also be studied.

Figure 4: Artist's impression of a geological disposal facility in Boom clay

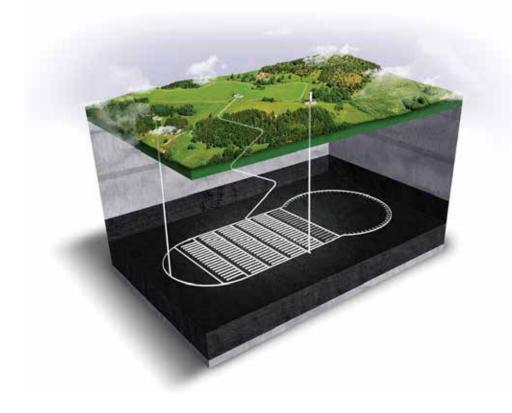


Figure 5: Artist's impression of OPERA supercontainer for heat-generating HLW



#### **Current status**

After two calls for proposals (in 2011 and 2012), the OPERA research programme comprises 21 projects that cover all (non-optional) tasks from the research plan. The first results will be expected in the second half of 2013.

Considering the broad group of researchers involved with contributions from almost 20 research institutes in the Netherlands and abroad and that the programme remained within budget, it can be concluded that the restart of geological disposal was successful. It can also be concluded that restarting research programme after ten years took a considerable effort, which justifies the focus of OPERA on continuing research after the five years the programme lasts and the development of a roadmap for the long-term research on geological disposal of radioactive waste in the Netherlands.

#### References

Commissie Opberging Radioactief Afval (CORA) (2001), Retrievable Disposal of Radioactive Waste in the Netherlands (Terugneembare berging een begaanbaar pad), Ministerie van Economische Zaken, Den Haag, Chapter 5 and page 24/110, February 2001 (Summary in English).

Dutch Government (1993), Cabinet Position on Deep Underground Disposal (Kabinetsstandpunt over de vraag of de diepe ondergrond mag en kan worden gebruikt voor het opbergen van afval), NMP-Action 62, 27 April 1993.

Heijdra, J.J. and J. Prij (1997), Concept ontwerp terughaalbare berging, ECN-C—96-087.

Humbeeck, H. Van, Chr. De Bock, W. Bastiaens (2007), "Demonstrating the Construction and Backfilling Feasibility of the Supercontainer Design for HLW", RepoSafe – International Conference on Radioactive Waste Disposal in Geological Formations, Braunschweig, Germany, November 2007, Paper 2B.14.

Ministry of Economic Affairs, Agriculture and Innovation & Ministry of Foreign Affairs (EL&I) (2001), National Report of the Kingdom of the Netherlands for the Fourth Review Conference on the Joint Convention of the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management, 1-148.

- Ministry of Housing, Physical Planning and Environment (VROM) (1984), Radioactive Waste Policy in the Netherlands; An Outline of the Government's Position, September.
- VROM (2002), Radioactive Waste Management; Policy Perspective 2002-2010 (Beleidsstandpunt onderzoek radioactief afval), Letter to the Lower House 28674, 11 November 2002.
- VROM (2001), 4th National Environmental Policy Plan (NMP), Summary 13 June 2001.
- Poley, A.D. (2000), Concept ontwerp van terughaalbare opslag van radioactief afval in diepe boorgaten in steenzout (Torad-B), NRG Petten.

# NEA/RWM/R(2013)10

# The current status of a safety case for heat-generating radioactive waste disposal in salt in Germany

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In 2010 new safety requirements for heat-generating waste were implemented in Germany. The objectives of the project "Preliminary Safety Analysis of the Gorleben Site" (VSG), which was started in 2010, are to: i) compile and review the available exploration data of the Gorleben site and knowledge on disposal in salt rock; ii) assess if repository concepts in rock salt can comply with the new requirements based on currently available knowledge; iii) evaluate if methodological approaches can be used for a future site selection procedure and which technological and conceptual considerations can be transferred to other geological situations. The current status and results of this study are presented.

#### Introduction

The current German disposal programme for heat-generating radioactive waste is characterised by the implementation of new safety requirements and – particularly for rock salt – by the Preliminary Safety Analysis of the Gorleben Site (VSG).

In 2010 the safety requirements governing the final disposal of heat-generating radioactive waste in Germany were implemented by the Federal Ministry of Environment, Natural Conservation and Nuclear Safety (BMU, 2010). The Ministry considers as its fundamental objective the protection of man and environment against the hazards from radioactive waste. Unreasonable burdens and obligations for future generations shall be avoided. The main safety principles are concentration and inclusion of radioactive and other pollutants in a containment-providing rock zone. Any release of radionuclides may increase the risk for men and the environment only negligibly compared to natural radiation exposure. No intervention or maintenance work shall be necessary in the post-closure phase. The retrieval/recovering of the waste shall be possible up to 500 years after closure.

The possibility of using the salt dome Gorleben as a repository for high active waste has been in discussion in Germany since the 1970s. Since then, scientists have been exploring the salt dome from above and below ground. The objective of the project preliminary safety analysis of the Gorleben site (VSG), which was started in 2010, is to assess if repository concepts at the Gorleben site or other sites with a comparable geology could comply with the above-mentioned requirements based on currently available knowledge (Fischer-Appelt, et al., 2013). It shall further be assessed if methodological approaches can be used for a future site selection procedure and which technological and conceptual considerations can be transferred to other geological situations. The objective includes the compilation and review of the available exploration data of the Gorleben site and on disposal in salt rock, the development of repository designs, and the identification of needs for future R&D work and further site investigations.

As the VSG requires special expertise, various project partners have teamed up with GRS. These include the Federal Institute for Geosciences and Natural Resources (BGR),

DBE Technology GmbH, the Institute for Mineral and Waste Processing, Landfill Technology and Geomechanics at TU Clausthal, the Institute of Disposal Research at TU Clausthal, the Institute for Rock Mechanics GmbH (IfG), the Institute for Safety Technology (ISTec), Karlsruhe Institute for Technology/Institute for Nuclear Waste Disposal (KIT/INE), International Nuclear Safety Engineering GmbH (NSE), and Dr. Baltes.

#### Preliminary safety analysis of the Gorleben site

#### Structure of the VSG

The VSG comprises of four consecutive stages (see Figure 1):

- 1. Fundamentals: This topic includes the description of the geological site and its future evolution over one million years; further, an inventory of the waste that could presumably be emplaced in a repository at the Gorleben site according to the current situation in Germany with its phase-out of nuclear energy (June 2011), and, finally, a generation of a concept to accomplish radiological safety and to demonstrate its compliance to the safety requirements (BMU, 2010).
- 2. Based on these fundamentals, repository concepts are developed aiming on operational safety, long-term safety and retrieval/recovering of the waste. Two emplacement variants, namely storage of spent fuels in drifts or in boreholes, and different types of canisters (POLLUX®, CASTOR®, BSK3R) and one optional variant emplacement for non-heat-generating waste are projected.
- 3. The analysis of the repository system is based on these concepts. The features, events and processes are compiled and described. They are used to derive scenarios and to assess the probability of the evolution of the system. Geomechanical analyses investigate the integrity of the geological barrier (containment-providing rock zone) for 1 million years considering external and internal events and processes such as glaciation, decay heat or gas generation. Similarly, the seals for shafts and drifts are designed and analysed. The radiological consequences are analysed by numerical models for the transport of the liquid and gas phase (two-phase transport) in the long-term safety analysis.
- 4. The feasibility of the repository system as a containment system for radionuclides and the methodology to show compliance with the safety requirements are assessed. The uncertainties, which e.g. result from the incomplete geological exploration of the Gorleben site and which require additional R&D, are shown (Beuth, et al., 2013a). The methodological approach shall be discussed for its suitability to compare repository sites and its technical transferability to repository sites in other geological formations.

#### Waste

The heat-generating radioactive waste will be composed of irradiated fuel elements from power reactors, vitrified reprocessing waste and irradiated fuel elements from research and prototype reactors (Peiffer, et al., 2011). As an option negligible heat generating waste is also considered to be disposed to assess the feasibility of the joint disposal in a separate area of the repository. A hypothetical amount and composition of this waste is assumed. It includes depleted uranium tails from enrichment (about 35 000 m³), graphite (about 1 000 m³) and mixed waste (about 15 000 m³).

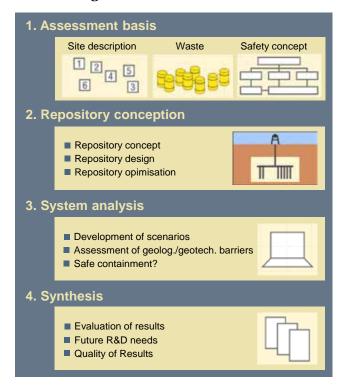


Figure 1: Structure of the VSG

#### Safety concept

In due consideration of the German safety requirements (BMU, 2010) the safety concept for the VSG project is based on the following principles:

- The radioactive waste must be contained in a containment-providing rock zone (CRZ), i.e. a part of the host rock enclosing the repository jointly with geotechnical barriers.
- · The containment shall be effective immediately after closure.
- · The containment must be provided by the repository system permanently and maintenance-free.
- · The intrusion of brine to the waste forms shall be prevented or limited.

According to the safety requirements a site for disposal of heat-generating radioactive waste is only suitable if a sufficiently large containment-providing rock zone is available. Its integrity must be ensured for 1 million years and a robust, staggered, maintenance-free multi-barrier system from technical components (container, drift seal, shaft seal,...) must be developed, which prevents an unacceptable release of radionuclides over the short and long term. This principle is also applicable if a barrier fails partially. Safe containment has to be demonstrated for probable and less probable evolutions of the site, while evolutions with very low probability (less than 1% over the demonstration period of 1 million years) need not to be considered. Criticality must be excluded in all phases of repository development.

The evaluation of the safety of the system includes the assessment of the probabilities and the consequences according to the concept for safety and proof. The proof considers four steps and applies an indicator known as the Radiological Insignificancy Index (RII) (Figure 2). The total release of radionuclides from the containment-providing rock zone

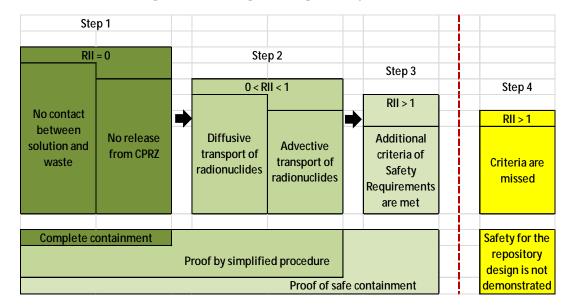


Figure 2: Radiological Insignficancy Index (RII)

and technical barriers is used in a generic model for radiation exposure. The RII is then calculated as a ratio to a radiation dose, which is considered insignificant (Mönig, et al., 2012).

- Complete containment is provided if there is no contact of the waste with solutions and no gaseous radionuclides are released from the containment-providing rock zone (RII = 0).
- The safe containment of radionuclides is achieved if the RII is greater than 0 and is less than 1. A simplified procedure is sufficient as proof. The assessment distinguishes between a release by diffusion or advection.
- · If the RII is greater than 1 additional criteria have to be met. Additional criteria refer to the individual radiation dose and are related to the probability of the evolution of the system (scenarios). A more detailed procedure is required.
- If these additional criteria are missed the designed repository is not feasible. Safe
  containment cannot be provided by the repository concept. The design of the
  repository has to be changed and assessed again. When all possible measures are
  optimised and safe containment still cannot be demonstrated, the site is not
  suitable.

#### **Uncertainties and assumptions**

There are some uncertainties for the Gorleben site which cannot presently be reduced further due to the present status of knowledge or even eliminated, e.g.:

- · The total lateral size of the salt dome is not yet known.
- · The features of the salt rock are known only for the explored area.
- The extension of the Hauptsalz may not be large enough for all designed repository concepts including the required safety distance to adjacent rock layers.

Therefore assumptions have been made which should be verified in the future. The main assumptions are:

- The lateral size of the salt dome is in accordance with the geological sketch of Bornemann, et al. (2011).
- The known features from the salt rock currently under exploration can be extrapolated to the entire area necessary for the whole repository.
- The extension of the Hauptsalz is sufficiently large for all designed repository concepts including the required safety distance to adjacent rock layers.

Geotechnical measures shall provide long- and short-term barriers. The long-term barrier is the backfill with salt grit. Its initial high porosity and permeability is reduced continuously by compaction. This is a time-dependent process and re-establishes the features of the undisturbed rock salt within the lifetime of the short-term barriers.

The short-term barriers are drift and shaft seals. These are composed by layers of different material providing diversity and redundancy. The failure of a drift or shaft seal is regarded as a less probable scenario.

Additional uncertainties concern data, parameters, and models. These are dealt with in deterministic model calculations using bandwidths.

#### Repository concepts

The repository concepts for the Gorleben site are described for three emplacement variants:

- · Variant A: As an option, non-heat-generating radioactive waste is emplaced in a separate area of the repository. This variant is combined with the following variants for spent fuel (Figure 3).
- · Variant B: Emplacement of heat-generating radioactive waste (spent fuel and vitrified waste) in self-shielding waste containers (POLLUXã casks) in horizontal drifts (Figure 3). As an alternative, the emplacement of heat-generating radioactive waste in transport and storage casks (CASTORã) in horizontal boreholes is considered although this requires an enhanced technical design for shafts and underground transportation.
- · Variant C: Emplacement of heat-generating radioactive waste in multi-purpose conical overpacks (Figure 4) in deep vertical boreholes.

The overall layout is optimised to minimise the size of the repository as well as to comply with temperature criteria. The technical installation and casks/containers are selected to ensure manageability, radiation protection, and operational safety. Disposal is planned using retreat working.

#### Scenario analysis

The site and the repository system will undergo exactly one evolution, which will be governed both by climatic and geological processes at the site and processes induced by the repository construction and the emplacement of heat-generating waste. This evolution cannot be predicted in all details.

A novel scenario development methodology is developed in the project VSG (Beuth, et al., 2013a). It aims at deriving one reference scenario for each repository design (horizontal drift/borehole emplacement) and a number of differing alternative scenarios. At large, the scenarios shall comprehensively represent the range of possible repository system evolutions. The methodology allows straightforward assignment of probability classes to the scenarios according to the regulatory framework (BMU, 2010). The individual

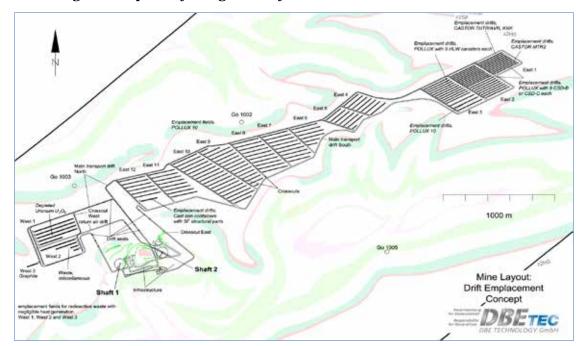
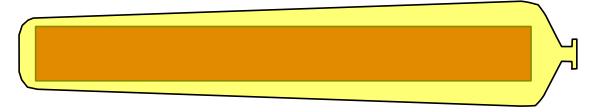


Figure 3: Repository design and layout (combination of Variants A and B)

Figure 4: Conical overpack (Variant C)



scenarios are described by features, events, and processes (FEP) that determine the future evolution of the final repository system at Gorleben (Wolf, et al., 2013). FEP may initiate or influence other FEP, be influenced by or result from other FEP. These interdependencies are used to derive scenarios systematically. The reference scenarios are derived from probable FEP and basic assumptions. The alternative scenarios are generated from violation of assumptions, from less probable FEP and from probable FEP with less probable parameter values. The human intrusion scenario is reviewed separately (Beuth, et al., 2013b).

# System analysis

The system analysis addresses the general questions:

- · Will the integrity of the geological salt barrier remain intact under the expected loads, e.g. like heat generation or glaciation?
- · Is there any flow of brine to the emplacement areas?
- · Are radionuclides released from the containment-providing rock zone?
- If so, what radiological consequences have to be expected?

Based on these scenarios an analysis of geomechanical integrity is performed. A demonstration of integrity is required for probable scenarios (BMU, 2010). The integrity has to be checked for less probable scenarios and their radiological consequences have to be analysed. The final step is the assessment and synthesis of the results.

The dilatancy criterion and the fluid pressure criterion are the main criteria to assess the geomechanical integrity of the rock salt barrier. The dilatancy criterion specifies that no damage to the rock fabric (e.g. induced cracking or interlinking of intercrystalline pore space) must occur in response to deviatoric stresses. The damage process is associated with dilatancy, i.e. an increase in volume caused by the development of micro-cracks and crack accumulations.

The fluid pressure criterion specifies that the smallest formation stress (considering compressive stresses as positive) in the barrier, plus any tensile strength which may be present, must be larger than the fluid pressure at the given depth. If this criterion is satisfied, fracturing of the host rock by fluid-pressure-driven penetration of fluids into the rock can be excluded.

The integrity of the salt barrier is only ensured if both criteria are satisfied in a sufficiently large zone around the underground workings of the repository. Linked flow paths from the water-bearing horizons in the overburden down to the emplacement zone, as well as release of hazardous substances from the repository itself (e.g. due to generation of a gas pressure) can then be excluded from a geomechanical point of view.

From the mechanical and thermo-mechanical simulations using a range of codes and material laws the following results and conclusions are obtainable:

- The emplacement of heat-generating waste heats up the salt dome over a large volume, but the thermally-induced stresses and deformations should not generate any continuous migration paths.
- The highest thermo-mechanical stresses affecting the salt barrier will occur within the first hundred years after sealing the geologic repository. Any loss of integrity of the barrier should become even less likely in the subsequent time period. Mechanical damage caused by exceeding the dilatancy limit should affect only the rock zones directly adjacent to the underground cavities and very distant rock zones.
- The thermo-mechanical stresses for the borehole emplacement design are expected to be higher than those for the drift emplacement concept because the heat is released in a smaller and differently shaped volume.

The integrity of the geotechnical barriers (drift and shaft seal) shall be demonstrated by numerical calculations concerning geological, thermal and geochemical impacts during their lifetime and by providing redundant and varying types of sealing systems in combination.

#### Long-term safety assessment

For the long-term safety assessment radionuclide transport will be modelled using a two-phase model, TOUGH2 (Pruess, et al., 1999), and a one-phase model, MARNIE (Martens, et al., 2002). The layout of the repository for the drift emplacement concept is transferred into a 3-D grid for TOUGH2 and a 1-D grid for MARNIE. This includes simplification steps due to constraints of the codes.

Even a failure of a single seal should not result in advective flow of brine within or into the emplacement fields. Furthermore the salt grit is expected to be compacted in a relatively short time when applying conservatively selected parameters according to experimental data for modelling of the compaction process.

A conservative assumption will be used for the final porosity of the salt grit after compaction. Using the 1-D grid no radionuclide transport by advection should be detected in the liquid phase beyond the containment-providing rock zone (CRZ). As a consequence any radionuclides in the liquid phase will be transported by diffusion only. The release of radionuclides will be calculated through the eastern drift seal (Figure 2, close to cross-cut east) and through the western seal.

Radionuclide transport and release via the gas phase from the CRZ may be relevant after closure and will be calculated in a two-phase model with TOUGH2. The compaction of salt grit and metal corrosion with gas generation will be driving forces on the transport and release of gaseous radionuclides (e.g. <sup>14</sup>C as methane or carbon dioxide from structural parts) through a drift seal.

- the compaction rate;
- the advective and diffusive transport parameters of radionuclides at low salt grit porosities;
- the diffusion coefficient in high compacted salt grit;
- · the solubility limits for some radionuclides;
- · the release of radionuclides into salt grit;
- · the formation of gaseous radionuclides.

#### Synthesis of the project results

The safety concept, which was generated during the course of the project, is suitable to demonstrate its compatibility with the safety requirements. The generated design of the repository system will be checked for its feasibility and compatibility with the safety requirements. Nevertheless some assumptions are necessary. The assumptions refer to the status of geological exploration, the reliability of construction and some inherent uncertainties.

Optimising strategies regarding the repository design will be studied. These may include a changed repository layout, implementing a void volume as a sink (e.g. an infrastructure area backfilled with gravel), hindering any gas flow or use of gas tight casks to confine volatile radionuclides.

Conclusions will be provided on:

- the possible release of gaseous radionuclides, the two-phase flow processes and the subsequent model for radiation;
- the minimisation of the containment-providing rock zone;
- the handling of combinations of less probable but interdependent FEP;
- the requirements for other pollutants and the heating of groundwater;
- repeating a safety analysis;
- · indications if a repository in salt rock is feasible.

#### Applicability to a site selection process

During the course of the project the request emerged in politics to establish a transparent, stepwise site selection process in Germany. Alternative sites should be identified and explored in addition to the Gorleben site. This procedure foresees a

development of site selection criteria and standards for site comparison, which should be developed for different steps of the process. Safety analyses are foreseen to evaluate the results of surface and subsurface explorations of possible sites.

The results of the VSG will be valid for a repository site in salt rock. The methodology may be transferred to sites in other geological formations.

#### References

- Beuth, T., et al. (2013a), "Szenarienentwicklung: Methodik und Anwendung", Bericht zum Arbeitspaket 8, GRS-284, ISBN 978-3-939355-60-1, Gesellschaft für Anlagen- und Reaktor-sicherheit (GRS) mbH, Köln.
- Beuth, T., et al. (2013b), "Untersuchungen zum menschlichen Eindringen in ein Endlager", Bericht zum Arbeitspaket 11, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-280, ISBN 978-3-939355-56-4, Gesellschaft für Anlagen- und Reaktor-sicherheit (GRS) mbH, Köln.
- Bornemann, O., et al. (2011), "Description of the Gorleben Site, Part 3: Results of the Geological Surface and Underground Exploration of the Salt Formation", Bundesanstalt für Geowissenschaften und Rohstoffe, Hannover, ISBN 978-3-9813373-6-5.
- Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU) (2010), Sicherheitsanforderungen an die Endlagerung wärmeentwickelnder radioaktiver Abfälle (Stand: 30.09.2010), BMU, Bonn, Germany.
- Fischer-Appelt, K., et al. (2013), "Synthesebericht für die VSG", Bericht zum Arbeitspaket 13, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-290, ISBN 978-3-939355-66-3, Gesellschaft für Anlagen- und Reaktor-sicherheit (GRS) mbH, Köln.
- Martens, K.-H., H. Fischer and P. Romstedt (2002), Beschreibung des Rechenprogrammes MARNIE, 135 Seiten, GRS-A-3027, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Köln.
- Mönig, J., et al. (2012), "Sicherheits- und Nachweiskonzept", Bericht zum Arbeitspaket 4, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-277, ISBN: 978-3-939355-53-3, Gesellschaft für Anlagen- und Reaktor-sicherheit (GRS) mbH, Köln.
- Peiffer, F., et al. (2011), "Abfallspezifikation und Mengengerüst", Bericht zum Arbeitspaket 3, GRS-278, ISBN 978-3-939355-54-0, Gesellschaft für Anlagen- und Reaktor-sicherheit (GRS) mbH, Köln.
- Pruess, K.O. and G. Moridis (1999), TOUGH2 User's Guide, Version 2.0, Earth Sciences Division, Lawrence Berkeley National Laboratory, LBNL-43134, University of California, Berkeley, California, USA.
- Wolf J., et al. (2013), "FEP-Katalog für die VSG Konzept und Aufbau", Bericht zum Arbeitspaket 7, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-282, ISBN 978-3-939355-58-8, Gesellschaft für Anlagen- und Reaktor-sicherheit (GRS) mbH, Köln.

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# Session III Integrity of Rock Sal

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# NEA/RWM/R(2013)10

# Applicability of natural analogues for a safety case in salt

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Natural analogue studies represent an important category of supporting arguments for the safety case. The main benefit of natural analogues in a safety case is to increase the understanding of the full complexity of long-term processes. In order to use this potential benefit of natural analogues for a SC a systematic review of the required key information for the safety assessment has to be carried out. This contribution summarises the results from a German R&D project. This project aims at identifying the key information for a HLW repository in salt. One important question is how this information may best be fulfilled by natural analogue studies. Therefore the natural analogues are categorised to illustrate how they contribute to the important elements of the safety assessment in salt. In addition, the status of the natural analogues is assessed regarding their role for a safety case in salt in Germany.

#### Natural analogues in the safety case

Before a repository for radioactive waste can be operated, its safety must be adequately evaluated. The iterative process of evaluating the safety of a repository is known as a safety case (SC), and requires detailed analysis of the repository system and its evolution. One important aspect of the SC is that its safety statement and its robustness rely on multiple lines of arguments. Diverse sources of information are brought together to form a consistent picture of the characteristics and history of a site, from which a reliable prognosis of future evolution can be made (NEA, 2004).

One important source of information is the study of natural analogues, i.e. the investigation of natural, anthropogenic, archaeological or industrial systems which have some definable similarity with a radioactive waste repository and its surrounding environment (Miller, et al., 2006). As in this definition all natural, human and technical analogues are subsumed under the term "natural analogues" (NA).

The main value of NA studies is to provide information on the full complexity of the repository system and on the characteristics of processes over long time scales. In general, the direct use of quantitative information from NA studies in SC is limited, since it is very difficult to extract hard numerical data from complex natural systems where initial and boundary conditions are afflicted by some degree of uncertainty. Nevertheless, the use of NA is very useful to support laboratory and in situ data and to test numerical codes used in the SC. Furthermore, NA are observable, tangible and understandable to the lay stakeholder, providing valuable information in the public eye. They are an important tool for technical and public confidence building.

#### Studies and activities on natural analogues in salt

There is already very comprehensive literature on the use of NA for the SC with a focus on repositories in clay and granite (e.g. Chapman, et al., 1984; Miller, et al., 1994, 2000).

Although there have been activities regarding NA in salt for more than twenty years, there are only a few published NA studies that addresses salt-specific aspects (e.g. Knipping, 1989; Brenner, et al., 1999). Being aware of this situation, Noseck, et al. (2008) described NA studies performed for high-level waste (HLW) repositories in salt in Germany.

In 2005, the German research project ISIBEL was started to develop a novel approach to prove the safety of a HLW repository in a salt formation, to refine the safety concept, to identify open scientific issues and to define necessary R&D work. The yardstick for the work is the ability to develop a SC according to the state of the art. The project is funded by the German Ministry of Economics and Technology (BMWi) and it is concertedly executed by DBE Technology GmbH, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR) and Gesellschaft für Anlagen- und Reaktorsicherheit (GRS). The first project phase covered October 2005 to March 2010. In this phase several open issues were identified for the German SC. The follow-up project phase started in 2010 and addresses several of the identified open issues. One issue is the applicability of NA for the German SC. The assessment of the applicability of NA requires a systematic review of the key information required for the SC. The results of this review in the ISIBEL project are presented in this contribution.

#### Required key information for a safety case in Germany

For the disposal of HLW in rock salt the geological barrier is of utmost importance. Due to the German Safety Requirements (BMU, 2010) the radioactive waste must be contained in a defined rock zone in such a way that it remains in situ and, at the most, only a small quantity of material may leave this rock zone, the so-called containment-providing rock zone (CRZ). The containment is thus mainly accomplished by the geological barrier. Due to the inevitable penetration of the geological barrier by the construction of the mine, a technical barrier system is required to seal the CRZ.

Figure 1 shows the key elements of a SC according to the approach developed in the project ISIBEL. Owing to the uncertainty in predicting the real evolution of the site, plausible scenarios have to be developed. This is carried out systematically on the basis

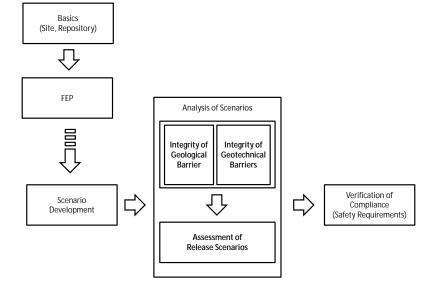


Figure 1: Key elements of the German safety case

of FEP catalogue. According to the Safety Requirements the containment has to be demonstrated for probable and less probable developments (scenarios) of the site over

the demonstration period of 1 million years. To verify that the safety criteria of the Safety Requirements are met, the scenarios have to be analysed. Scenarios with very low probability (less than 1%) can be neglected in the analysis.

The analysis of the scenarios comprises three main parts:

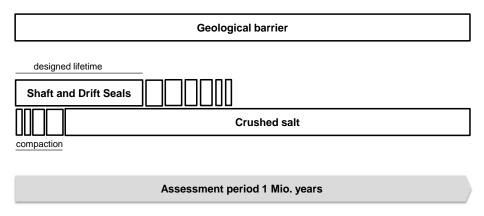
- The demonstration of the integrity of the geological barrier is primarily based on the results obtained from mechanical and thermo-mechanical model calculations, which describe the physical processes in the rock that are to be expected in the long term. In addition to the numerical calculations, geological and geo-engineering explorations, laboratory analyses of petrophysical properties, geotechnical in situ measurements and surveys, on-site observations, and mining experience from comparable geologic media and mines are also taken into account. The functionality and integrity of the salt barrier is considered to be mathematically proven if the formation of pathways can be excluded from a geomechanical point of view. Two criteria are applied:
  - Dilatancy criterion: The integrity of the barrier is ensured if no damage in the rock mass occurs. Comprehensive laboratory analyses of the petrophysical properties of salt showed that stress states below the dilatancy limit do not cause any damages, not even in the long term, which means that for stress states in this range, the impermeability of the rock is ensured. Only stress states above the dilatancy limit cause a loosening of the microstructure, which in turn leads to the gradual formation of a network of microfissures if such conditions persist.
  - Brine pressure criterion: Integrity is ensured if the lowest main compressive stress does not drop below the value of the hydrostatic pressure to be assumed at the corresponding depth. This pressure is calculated from a hypothetical column of liquid that extends to the surface.
- The demonstration of the integrity of the geotechnical barriers comprises the geotechnical barriers that are according to the safety concept of highest importance for the containment of the waste. In the safety concept developed in ISIBEL the safety concept takes credit from the drift and shaft seals. The demonstration of their integrity involves the consideration of the integrity of the barrier structure itself and of the hydraulic properties and their corresponding evolution over time of the barrier structure, the excavation damage zone (EDZ) and the contact zone between both. The requirements on the hydraulic resistance of the barriers are determined by hydraulic calculations carried out in the long-term safety analysis. The proof of the integrity of the barrier structure is based on the demonstration and documentation of:
  - structural and mechanical integrity;
  - crack restriction;
  - long-term stability.

Furthermore, their producibility has to be demonstrated. Possible impacts on the barrier structure, as derived in the scenario development, have to be taken into account. The requirements on the hydraulic resistance as well as on the design life of the shaft and drift seals are directly correlated with the compaction behaviour of the crushed salt backfill material. In the long run the compacted crushed salt takes over the containment function of the shaft and drift seals (Figure 2). Therefore the understanding of the compaction behaviour of crushed salt is of high importance.

The assessment of release scenarios complements the demonstration of the integrity
of the geological and the geotechnical barriers. In case of scenarios for which the
integrity of the geological barrier or the geotechnical barrier cannot be proven,
containment within the CRZ is possibly not given. The consequences arising from
such scenarios must be analysed.

Finally, a verification of all scenarios if the safety criteria of the Safety Requirements are met is carried out.

Figure 2: Functional requirements for the barrier system (not to scale)



#### Review and assessment of natural analogues

The main objective of the work is to carry out a compilation and evaluation of how NA can be used for the safety case of a HLW repository in salt. The assessment is structured to answer the following questions:

- · For which aspects can NA contribute to the assessment of safety?
- · What is the status of the identified NA?
- · How does the NA contribute to the confidence building of the SC (communicability)?

The first question is answered by starting a systematic review of the required key information needs of the SC and an assessment if these requirements can be fulfilled by NA information. In order to achieve this, the compiled NA are divided in three types of NA in order to highlight the important elements of the safety and assessment concept:

- 1. NA for the integrity of the geological barrier;
- 2. NA for the integrity of the geotechnical barriers;
- 3. NA for release scenarios.

In order to answer the second and third question, the assessment considers two different schemes. The first scheme assesses the status of NA for a SC in salt. Five classes are chosen for this assessment:

- · NA is identified and documented.
- · NA is identified and needs to be better documented.
- NA is identified (no documentation).
- NA is not identified.
- · NA is (probably) not identifiable.

The second scheme assesses the communicability of a NA for the SC in salt. Two classes are chosen for the assessment:

- · Public confidence building: NA is tangible and understandable to the lay stakeholder.
- Technical confidence building: NA is an argument to support the understanding of complex behaviour.

In the following, studies are compiled and assessed according to these schemes for the three defined types of NA that are of importance for the SC in salt (Tables 1.1 to 1.3). If the status is assessed as ++, the documentation is cited. In order to limit the number of references other documentations are generally not referenced. More literature can be found in the cited overview articles. It should be noted that the work done in the research project ISIBEL contains only the identification of a potential study and an evaluation if these studies could be relevant generally for supporting a SC in salt. For a specific site, these potential analogues need to be critically evaluated to ensure their appropriateness to this site.

#### Analogues for the geological barrier

In total, 11 aspects have been identified that could contribute as NA to a SC in salt for the integrity of the geological barrier (Table 1). For the assessment it is distinguished between the situation at the site (self-analogue, here the salt dome Gorleben is used exemplarily) and NA at other sites.

Table 1: Compilation and assessment of NA for the geological barrier (status of assessment: 2010/11)

No.	Aspect/study	Applicability in SC (key information)	Status site	Status NA	Confidence building
1	Occurrence of salt domes	Long-term stability of salt domes	++	+	00
2	Neotectonic conditions	Occurrence of earthquakes and magmatic events	++		00
3	Analysis of the salt flow	Uplift rates	++	+	00
4	Thickness and composition of the cap rock	Subrosion rates	++	+	0
5	Behaviour of competent salt formations	Possible water pathways	+	•	0
6	Br (and Rb) distribution in minerals	Interaction between formation and external solutions	++	++	0
7	Chemical and isotope composition of fluid inclusions	Interaction between formation and external solutions and gases	++	++	0
8	Openings from salt mining	Behaviour of salt at disposal level	•	+	0
9	Basalt intrusions	Behaviour of salt at high temperatures		++	00
10	Basalt intrusions	Sealing of fissures		++	0
11	Cryogenic fractures	Occurrences of fractures formed by salt contraction during cooling	-	+	0

- ++ NA is identified and documented
- + NA is identified and need to be better documented
- NA is identified (no documentation)
- NA is not identified
- -- NA is (probably) not identifiable

All identified NA are related to the containment-providing properties of the geological barrier and are explained in the following enumeration:

- 1. The fact that more than 250 million years after sedimentation of the evaporites a large number of salt domes exist in the world, e.g. in the Permian North German Plain, provides arguments for the integrity of salt rock, the slow dissolution of salt in the deeper underground and its impermeability and mechanical and structural stability over geological time frames. There are several documentations of salt formations, e.g. for the Gorleben salt dome (Bornemann, et al. 2008). This fact is a tangible and understandable argument for the lay stakeholder and should be used as a general argument for the advantages of the host rock salt.
- 2. The assessment of neotectonic condition is an important argument for the prognosis of the future evolution of the repository system (uplift or subsidence of the area, expected intensity of earthquakes, occurrence of magmatic events) and is expected to be very important for the discussion with the lay stakeholder. This can be described for the investigated site, e.g. Köthe, et al. (2007), but no further NA can be applied, since the condition cannot be transferred. This aspect cannot be supported by NA.
- 3. For the future evolution of a salt dome it is of utmost importance to be able to predict its uplift (stage of diapirism). Other important processes depend on the expected uplift rates, e.g. subrosion (see below). Valuable information can be drawn by the analysis of salt flow in the past and remaining salt in the catchment area of the salt dome. An example is the description of development of the Gorleben salt dome by Jaritz (1994) and Zirngast (1991). A comparison with other salt domes provides a deeper understanding of the process and supports the prediction of future uplift rates. This is an aspect that can be also comprehensibly illustrated for the public.
- 4. An important process is the salt dissolution from the top of the salt surface which might lay open the repository area over very long time scales. During the subrosion process low solubility minerals such as anhydride, gypsum or clayey material become enriched (cap rock). Dating of the recently built cap rock layer is a good argument for the derivation of subrosion rates. Examples for the Gorleben analyses of cap rock layers of salt domes and derived subrosion rates are given in Köthe, et al. (2007). As for the uplift rates a comparison with other salt domes provides a deeper understanding of the process and supports the prediction of future rates.
- 5. The fact that anhydrite is a very common rock in a salt formation, which behaves in a stiff and brittle manner and can provide a possible pathway for fluids is a very important aspect for a SC in a salt dome. Competent layers (anhydrite) embedded in incompetent material (rock salt) break under mechanical stress. Several lab investigations and the results from the Gorleben exploration area show this behaviour. This should be investigated and documented at other salt formations, especially salt domes, to support these important observations.
- 6 and 7. Concerning the integrity of the salt dome, arguments demonstrating that external fluids from adjacent or overlying strata did not migrate into the interior of the salt dome are of great importance. It is a strong argument that no fluid pathways existed in the formation. NA studies like profiles of content and isotope signatures of gases in fluid inclusions can be used to support such conclusions. For the salt formations in the Permian North German Plain several detailed studies are available, e.g. Hermann, et al., (1991a, 1991b), Siemann & Ellendorf (2001), Potter, et al. (2004). A second aspect to support this argument is the interpretation of bromide distribution in the evaporites. If the characteristic bromide profile of the evaporation phase is preserved, it can be followed that there was no interaction with fluids after sedimentation, e.g. Schulze (1960), Braitsch (1963a, 1963b), Becker (2008). In general, the understanding of such interpretations requires an advanced knowledge in geochemistry and is rather appropriate for the discussion of experts.

- 8. The mechanical behaviour (especially deformation processes) of salt in several hundreds of meters depth could be analysed by investigations of existing and former salt mines. Despite the vast activities of the salt mining and gas storage industry it has been analysed only in a few studies, how far NA can support the understanding of such mechanical processes, e.g. Brenner, et al. (1999).
- 9 and 10. Thermal stability is an important aspect due to the heat production of HLW. The geologic analogue of salt penetrated by volcanic dikes can be used to demonstrate that transient high-temperature processes have a very limited influence on the salt structure (Knipping, 1989).
- 11. NA can also help in controversial discussions. For example, in the discussion on the origin of joints (up to 600 m depth) in the salt dome Bokeloh near Hannover, Germany, Bauer (1991) suggests that the low temperatures in past glacial periods could be one reason for the evolution of these joints. Other reasons, e.g. a tectonic origin, are also plausible. Here, investigations of other domal salt structures can support the discussion.

### Analogues for the geotechnical barriers

According to the concept of demonstrating the integrity of the geotechnical barriers it is important to analyse the structural and mechanical (including hydraulic) behaviour and the long-term stability of these barriers. NA can support this analysis.

Table 2: Compilation and assessment of NA for the geotechnical barriers (Status of assessment: 2010/11)

No.	Aspect/study	Applicability in SC (key information)		Confidence building
1	Investigations of bulkhead drift	Reduction of the permeability of an EDZ around drift sealing	+	0
2	Basalt intrusions	Long-term behaviour of basaltic gravel	++	00
3	Chemical and mineralogical composition of natural clays	Impact of high temperatures on clay minerals	++	0
4	Properties of natural salt clays in salt	Long-term behaviour of clays as sealing material	+	0
5	Corrosion of historical concrete buildings	Long-term behaviour of cementitious materials	++	0
6	Bentonites in saline environment	Long-term behaviour of bentonite as sealing material	+	0
7	Compacted backfill material from old drifts in salt mines	Compaction of crushed salt over long time scales	•	0

- ++ NA is identified and documented
- + NA is identified and need to be better documented
- NA is identified (no documentation)
- NA is not identified
- -- NA is (probably) not identifiable

Table 2 contains all identified NA for the integrity of the geotechnical barriers:

- 1. The excavation of the repository infrastructure with drifts and boreholes changes the favourable properties of the rock salt amongst others by an increase of permeability in a specific zone around the voids. NA giving evidence that healing, i.e. decrease of the permeability back to that of the undisturbed rock salt occurs are of great importance for the safety case and support the numerical models describing the time-dependent decrease of permeability in the EDZ around the seal structures. As an example for short-term processes the permeability reduction of the excavation disturbed zone (EDZ) in rock salt can be observed in old drifts (Bechtold, et al., 2004; Wieczorek, et al., 2001).
- 2. to 6. A very important field where NA can provide valuable information is the long-term stability of the materials used in a repository (waste forms, waste packaging, buffers, backfills and seals). NA can provide information on the long-term behaviour of this (or homologues) materials complementarily to laboratory investigations. In the salt concept different cementitous and bentonite are used in the seal structures. Several studies are available (Jull, et al., 1990; Roy, et al., 1982, 1983; Pellegrini, et al., 1999). For basaltic gravel (one option for filling parts of the shaft column) Knipping (1989) can be used to get valuable information for its long-term behaviour in salt.
- 7. As explained above, the compaction of crushed salt plays a decisive role in the safety concept (Figure 2). This process is important for time scales that cannot be covered with laboratory experiments. Therefore it is very important to find NA that confirm the understanding gained from these experiments. One option is the investigation of old backfilled mines. A first analysis of possible activities was carried out in Brenner, et al. (1999), but more effort is necessary to document NA for this important aspect of the German safety case.

#### **Analogues for release scenarios**

Although containment (that means the integrity of the geological barrier and the geotechnical barriers) is the first priority in the German Safety Concept, retardation processes provide additional valuable safety functions and should be supported by NA. The identified NA for release scenarios are listed in Table 3:

- 1. and 2. The waste matrix is a barrier not taken credit from in the safety concept. But nevertheless it has an important role by retarding the release of radionuclides from the waste.
  - 3. and 4. Retardation of radionuclides, sorption.
- 8. and 9. In case of radionuclide release into the overburden, the quaternary and tertiary sediments of the overburden represent an additional barrier for radionuclide transport. In order to understand the long-term behaviour of radionuclides in such systems, an analogue site in the Czech Republic was studied, where uranium enrichment in argillaceous lignite-rich sediments occurred in a tertiary basin (Noseck and Brasser, 2006).

#### **Conclusions**

The use of NA is one important element for alternative lines of reasoning to complement the results of the safety assessment in SC (IAEA, 2012). But it is equally important that NA not be viewed in isolation. Their key role is to be complementary to laboratory studies and modelling exercises and to increase the understanding of complex long-term processes. In order to use this potential benefit of natural analogues a systematic review of the key information required for a SC has been carried out in the ISIBEL project for a generic repository in a domal salt structure.

Table 3: Compilation and assessment of NA for release scenarios (status of assessment: 2010/11)

No.	Aspect in the SC	Applicability (key information)	Status NA	Confidence building
1	Stability of natural basaltic glass	Corrosion of borosilicate glass	+	0
2	Uraninite deposits	Corrosion of spent fuel	-	00
3	Basaltic glass in saline environment	Formation of secondary phases during glass corrosion and retardation of radionuclides	+	0
4	Co-precipitation and sorption of radionuclides	Retardation of radionuclides, e.g. on products from metal corrosion	+	0
5	Lanthanide distributions in low soluble mineral fractions of marine evaporites	Mobility of lanthanides (as chemical homologue) for actinides in salt formations	+	0
6	Precipitation of natural elements during formation and recrystallisation of salt deposits	Retardation of radionuclides by co- precipitation with salts	-	0
7	Behaviour of radionuclides in highly saline systems, e.g. sole of geothermic deep drillings	Retardation of radionuclides under high saline conditions	+	0
8	In situ sorption values in sedimentary formation	Confirmation of sorption values for the overburden	+	0
9	Uranium migration in sedimentary formations	Behaviour of uranium and thorium in tertiary sediments of the overburden	+	0

- ++ NA is identified and documented
- + NA is identified and need to be better documented
- NA is identified (no documentation)
- NA is not identified
- -- NA is (probably) not identifiable

The compilation is an open list (state 2010/2011) and needs to be permanently updated. Newer NA studies are not considered.

The list has several advantages:

- · Statement of the importance for the SC.
- · Overview on available NA, transparent way of illustrating the considered studies.
- · It initiates discussion on NA for a specific SC.
- The site should have in the end ++, otherwise it an identification of R&D needs.

The work done in this research project contains only the identification of a potential study or NA and an evaluation if they could be accepted as relevant analogues to support a SC. Of course these potential analogues need to be critically evaluated to ensure their appropriateness to a specific site.

They should highlight less quantitative evidence for safety, including evidence from natural analogues, may be more accessible, more convincing and of more interest to the public than the results of complex mathematical models. The point is clearly to determine the objectives and success criteria for future analogue studies so that they can be better planned.

#### References

- Bauer, J. (1991), "Kryogene Klüfte in norddeutschen Salzdiapiren?" Zbl. Geol. Paleont., 1/4, 1247-1261.
- Bechthold, W., et al. (2004), Backfilling and Sealing of Underground Repositories for Radioactive Waste in Salt (BAMBUS-II Project), Commission of the European Communities, EUR 20621 EN, Brussels.
- Becker, F. (2008), "Geology of the Salt Deposit at the Brine Field Urcuit Near Bayonne, France", Kali und Steinsalz Heft, 3, 2008, 36-45.
- BMU (2010), Safety Requirements Governing the Final Disposal of Heat-Generating Radioactive Waste, Federal Ministry for the Environment, Nature Conservation and Nuclear Safety, 30 September 2010.
- Bornemann, O., et al. (2008), Description of the Gorleben Site Part 3: Results of the Geological Surface and Underground Exploration of the Salt Formation, Bundesanstalt für Geowissenschaften und Rohstoffe, Hannover.
- Braitsch, O. and A.G. Herrmann, (1963a), "Zur Geochemie des Broms in salina-ren Sedimenten: Teil I: Experimentelle Bestimmung der Br-Verteilung in verschiedenen natürlichen Salzsystemen", Geochimica et Cosmochimica Acta, Volume 27, Issue 4, April 1963, 361-391.
- Braitsch, O. and A.G. Herrmann, (1963b), "Zur Geochemie des Broms in salina-ren Sedimenten Teil II: Die Bildungstemperaturen primärer Sylvin- und Carnallit-Gesteine", Geochimica et Cosmochimica Acta, Volume 28, Issue 7, July 1964, 1081-1109.
- Brenner, J., et al. (1999), Untersuchung von Altversatz als Analogon zur Konvergenz und Kompaktierung versetzter untertägiger Hohlräume im Salz über lange Zeiträume Phase I, GRS-Bericht 147.
- Chapman, N.A., I.G. McKinley and J.A.T. Smellie (1984), The Potential of Natural Analogues in Assessing Systems for Deep Disposal of High-Level Radioactive Waste, Nagra Technical Report NTB 85-41, Nagra, Baden, Switzerland.
- Herrmann, A.G. and C.Z. van Borstel (1991a), "The Composition and Origin of Fluid Inclusions in Zechstein Evaporites of Germany", N. Jb. Miner. Mh., H6, 263-269.
- Herrmann, A.G., et al. (1991b), "The Quantitative Analysis of Fluid Inclusions in Marine Evaporites", N. Jb. Miner. Mh., H1, 39-48.
- International Atomic Energy Agency (IAEA) (2012), The Safety Case and Safety Assessment for the Disposal of Radioactive Waste, IAEA Specific Safety Guide, No. SSG-23, Vienna.
- Jaritz W. (1994), "Die Entwicklungsgeschichte des Standortes Gorleben als natürliches Langzeitverhalten eines Barrierensystems", Z. dt. Geol. Ges., 145, 192-206.
- Jull, S.P. and T.P. Lees (1990), Studies of Historic Concrete, Final Report, EUR 12972 EN, Office for Official Publications of the European Communities, Luxembourg.
- Klinge, H., et al. (2007), Description of the Gorleben Site Part 1: Hydrogeology of the Overburden of the Gorleben Salt Dome, Bundesanstalt für Geowissenschaften und Rohstoffe, Hannover.

- Köthe, A., et al. (2007), Description of the Gorleben Site Part 2: Geology of the Overburden and Adjoining Rock of the Gorleben Salt Dome, Bundesanstalt für Geowissenschaften und Rohstoffe, Hannover.
- Knipping, B. (1989), "Basalt Intrusions in Evaporites", Lecture Notes in Earth Sciences, 24, Springer-Verlag.
- Miller, W.M., et al. (1994), "Natural Analogue Studies in the Geological Disposal of Radioactive Wastes", Studies in Environmental Sciences, 57, Elsevier, Amsterdam, the Netherlands.
- Miller, W.M., et al. (2000), The Geological Disposal of Radioactive Wastes and Natural Analogues, Pergamon, Amsterdam, the Netherlands.
- Miller, W.M., et al. (2006), NAnet Network to Review Natural Analogue Studies and Their Applications to Repository Safety Assessment and Public Communication, Synthesis Report, EUR 21919.
- Noseck, U. and T. Brasser (2006), Radionuclide Transport and Retention in Natural Rock Formations Ruprechtov Site, Gesellschaft für Anlagen und Reaktorsicherheit (GRS) mbH, GRS-218, Braunschweig.
- Organisation for Economic Co-operation and Development/NEA (OECD/NEA) (2004), Post Closure Safety Case for Geological Repositories. Nature and Purpose, OECD/NEA, Paris.
- Pellegrini, R., et al. (1999), Natural Analogues of the Thermo-Hydro-Chemical and Thermo-Hydro-Mechanical Response, EUR 19114, Luxembourg.
- Potter, J., M. Siemann and M. Tsypukov (2004), "Large Scale Isotopic Carbon Fractionations in a Zechstein Evaporite Deposit and the Generation of Extremely <sup>13</sup>C-Enriched Methane", Geology, 32 533-536.
- Roy, D.M. and C.A. Langton (1982), Longevity of Borehole and Shaft Sealing Materials: Characterization of Cement-Based Ancient Building Materials, Technical Report ONWI-202, Office of Nuclear Waste Isolation, Columbus.
- Roy, D.M. and C.A. Langton (1983), Characterization of Cement-Based Ancient Building Materials in Support of Repository Seal Materials Studies, Technical Report BMI/ONWI-523, Office of Nuclear Waste Isolation, Columbus.
- Schulze, G. (1960), "Stratigraphische und genetische Deutung der Bromid-verteilung in den mitteldeutschen Steinsalzlagern des Zechsteins", Freiberger Forschungshefte, Reihe C83, Freiberg.
- Siemann, M.G. and B. Ellendorf (2001), "The Composition of Gases in Fluid Inclusions of Late Permian (Zechstein) Marine Evaporites in Northern Germany", Chem. Geol., 173, 31-44.
- Wieczorek, K., P. Schwarzianeck and T. Rothfuchs (2001), "Self-Healing of Excavation-Disturbed Rocks in the Near Field of Underground Cavities Exemplary Measurements in Rock Salt and Interpretation of Preliminary Results", Proceedings of the Eurosafe Conference, Paris, 5-6 November 2001.
- Zirngast, M. (1991), "Die Entwicklungsgeschichte des Salzstocks Gorleben Ergebnis einer strukturgeologischen Bearbeitung", *Geol. Jb.*, A132, Hannover.

# NEA/RWM/R(2013)10

# Geologic analogues for hot waste and for radionuclide releases

#### Norbert T. Rempe Carlsbad, New Mexico, USA

#### Introduction

Nature and engineering experience offer several practical worse- or worst-case analogues to the interactions of heat-generating radioactive waste forms with host rocks in geologic repositories. Magmas of basaltic and kimberlitic composition penetrated Paleozoic salt deposits in the United States and Germany (likely elsewhere as well) without compromising their competency or waste-isolating properties. Such vigorous thermal and hydrothermal disturbances of the host rock resulted - aside from the igneous rock crystallised from the magma - in only narrow contact-metamorphosed aureoles at the intrusions' margins and immediately adjacent pressurised reservoirs of intercrystalline fluids (mostly CO<sub>2</sub>) that did not migrate farther than centimetres to a few meters (if at all) for tens to hundreds of million years. Igneous intrusions into rocks other than salt, magma lakes in volcano craters, underground nuclear detonations, fossil natural nuclear reactors, and maturation windows for petroleum complement the picture of only short-range and largely insignificant effects of significant heat sources on the surrounding host rock. We therefore have no persuasive evidence to expect that, especially with somewhat judicious spacing of heat-generating waste packages, a geologic repository will not isolate such waste forms forever in any practical sense of the word.

Because the fundamental purpose of geologic isolation is protection against injury from radiation, any discussion about analogues must take into account the natural variability of ionising background radiation. Current local and regional background depends largely on geological and hydrological factors, e.g. radon-rich bedrock and underground spaces, monazite sand beaches, and springs issuing from rocks rich in radioisotopes. Background radiation was much higher in geologic history, during which life started and evolved. Temporal and spatial background variations and the lack of statistically significant evidence for negative consequences of exposure to much-higher-than-average background radiation must be included in unbiased analogue studies and rational justifications for radiation protection standards.

#### **Discussion**

#### Effects of geologic heat sources

A presentation to the Blue Ribbon Commission on America's Nuclear Future (Garrick, 2010) included these arguments:

- Heat generated by the waste affects geochemical processes and the rates of degradation of engineered barriers.
- · Extensive modelling is necessary to predict repository performance.

To the contrary, analogues demonstrate:

- · Many heat sources have negligible effects on the confinement capability of host rock, based on observations of natural and engineered analogues.
- Worst-case analogues bound repository performance well within reasonable limits, rendering detailed modelling and complete understanding of miniature-scale processes (even if that were achievable) unnecessary.

#### Contact metamorphism

Except in areas of pervasive regional metamorphism, caused by massive igneous intrusives or deep subsidence of rocks into domains of very high temperature and pressure, alteration (contact metamorphic) zones in rocks adjacent to heat sources are fairly narrow. Specifically for magma intrusions into salt deposits, these minor effects have been well documented by US and German authors (Knipping, 1989; Jahne, et al., 1994; Kaeding, 1962; Hermann, et al., 1989; Hermann, et al., 1993; Broughton, 1950; Kpekpasse, 2006; Flynn, 1996; Rempe, 2011). Herrmann & Knipping (1993) summarised succinctly:

The basaltic melts had a temperature of  $1\,150\,^{\circ}\mathrm{C}$  and the evaporite rocks a temperature of about  $50\,^{\circ}\mathrm{C}$  before intrusion of the magma. The basaltic melts were accompanied by mobile constituents, which had a temperature of only one hundred to several hundred degrees Celsius. The study of the evaporite rocks clearly shows that the alteration of the rock salt by the fluid components only extends a few centimetres into the evaporite and away from the contact to the basalt. In contrast, the mobile components penetrated much deeper into the K-Mg mineral associations of the potash salt seams, producing a zone of alteration which is up to and over  $10\,\mathrm{m}$  wide.

Direct exposures abound in potash and salt mines but have curiously been hardly noticed or studied by the radioactive waste community. One good (or actually bad) example for this remarkable lack of interest and follow-up is the underground outcrop in the Unterbreizbach potash mine in the Werra district described by Jahne, et al. (1994). Despite the authors' invitation and urgent plea for prompt scientific follow-up, because such a location can be kept accessible for only a short time in a working mine, nothing more was done, and a great opportunity was likely missed. Another prominent example is a dike/salt contact Intrepid Potash exposed only a few years ago in the Carlsbad potash district that will likely remain accessible for quite some time but should be studied before its freshness and quality deteriorate.

#### Diagenesis, hydrothermal alteration and petroleum maturation

Diagenetic process temperatures of  $145\text{-}160^{\circ}\text{C}$  are not uncommon, and many authors draw the line between diagenesis and low-grade metamorphism at about  $200^{\circ}\text{C}$ . Deep wells produce toxic and combustible fluids (hydrocarbons) from >200°C hot reservoirs that have isolated these fluids for hundreds of millions of years (Borak, et al., 1981). Fossil hydrothermal systems indicate that mineral alteration resulting from the flow of hot fluids through fractures extends only a few centimetres from the fracture wall into the matrix (Simmons, et al., 2012). Hydrocarbons begin to generate oil between ~60-80°C and become over-mature between ~150-200°C. The oil window ranges from ~60-100°C, and the gas window from ~100-200°C (Machel, et al., 1995).

The surface temperature of used fuel canisters, by comparison, may range from ~140-240 for <100 years (Brady, et al., 2009). In light of the broad evidence that sedimentary rocks and their confinement capabilities do not appear to be significantly affected by temperatures in this range, serious concern about their effects on repository host rocks seems exaggerated or at least highly debatable.

#### Spatial and temporal variability in natural background radiation dose rates

Natural geologic sources are the ultimate origin of most total population dose. Current natural background radiation varies over two orders of magnitude, based principally on local geology or hydrology or combinations of both.

Historical and local variations in the radioactivity of geologic materials and the frequently significant contribution of geologic environments to the magnitude of natural background levels of ionising radiation are not widely or sufficiently appreciated. Background radiation levels much higher than today's natural average likely played a role in early organic evolution. Early heat production from the decay of U, Th and K is estimated five times greater than at present. The decay of short-lived radioactive isotopes such as <sup>129</sup>I, <sup>244</sup>Pu and possibly <sup>26</sup>Al likely caused even higher heat production in the early Earth. Radioactive heat generation as a percentage of Earth's present total heat outflow appears to be at least 40% (Plant, et al., 1999). That estimate compares reasonably well with recent calculations based on terrestrial neutrino flux measurements (KamLAND Collaboration, 2011).

#### Natural analogue checks for radiation protection standards

Protection against harmful effects of ionising radiation is the professed principal goal of geologic isolation of radioactive waste. Many analogue studies focused on geochemical and hydrological systems and their effectiveness in preventing migration of radionuclides into the biosphere in concentrations above regulatory limits. Still missing is an unbiased comparison of limits and standards with the spatially and Earth-historically quite variable magnitude of background radiation. The Linear No-Threshold paradigm deserves to be challenged in the context of analogues of naturally high background levels without evidence or proof of concomitant harm to organisms. Monazite sand beaches in India and Brazil, igneous and metamorphic terrains with little or no sedimentary veneer in Sweden, Finland, and elsewhere, and radon spas in Iran and Austria are at least as worthy of analogue consideration as Roman nails, 17th century ship wrecks and natural nuclear reactors. Professional credibility requires addressing and resolving non sequiturs and logical inconsistencies such as those demonstrated below (Hart, 2002).

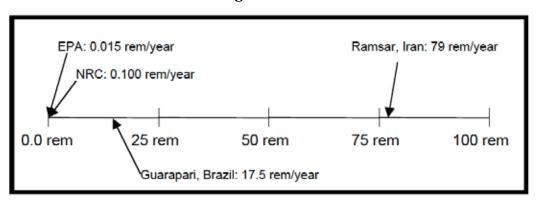


Figure 1: Scale comparing EPA and NRC regulatory limits to natural background radiation environments

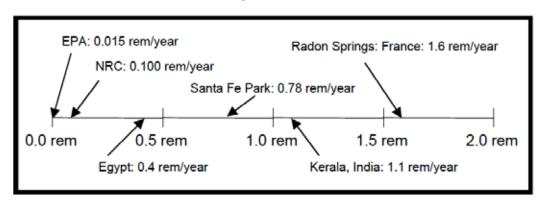


Figure 2: Expanded scale comparing EPA and NRC regulatory limits to natural background radiation environments

#### References

- Borak, B. and G.M. Friedman (1981), "Textures of Sandstones and Carbonate Rocks in the World's Deepest Wells (in Excess of 30 000 ft. or 9.1 km): Anadarko Basin, Oklahoma", Sedimentary Geology, 29, 133-151.
- Brady, P.V., et al. (2009), Deep Borehole Disposal of High-Level Radioactive Waste, Sandia Report SAND2009-4401.
- Broughton, J.G. (1950), "Observations on the Intrusion of Rock Salt by Peridotite", Transactions, American Geophysical Union, Volume 31, Number 2.
- Flynn, N.P. (1996), An Electron Microprobe Study of the Geothermometry of Some Kimberlite Dikes Cross-Cutting Coal, Salt and Shale, MS Thesis, Temple University, Philadelphia, PA.
- Garrick, B.J. (2012), "Experience Gained on the Management and Disposition of High-Activity Waste", Slide #7, Presentation to the Blue Ribbon Commission on America's Nuclear Future, Washington, DC, 16 November 2010, accessed 4 November 2012, http://cybercemetery.unt.edu/archive/brc/20120620235503/http://brc.gov/sites/default/files/meetings/presentations/garrickbrcslides111610.pdf.
- Hart, M.M. (2002), Disabling Radiological Dispersal Terror, Lawrence Livermore National Laboratory Report UCRL-JC-150849), accessed 14 November 2012, https://e-reports-ext.llnl.gov/pdf/246170.pdf.
- Herrmann, A.G. and B. Knipping (1989), Stoffbestand von Salzstöcken und Langzeit-sicherheit für Endlager radioaktiver Abfälle, PTB informiert, Clausthal-Zellerfeld.
- Herrmann, A.G. and B. Knipping (1993), "Waste Disposal and Evaporites", Lecture Notes in Earth Sciences, 45, Springer.
- Jahne, H. and D. Pader (1994), "Untertageaufschluß eines Basaltschlotes durch den Bergbau im Werra-Kaligebiet", Kali und Steinsalz, Band 11, Heft 7, September 1994.

- Kaeding, K.C. (1962), Geologische, magnetische und petrographische Untersuchungen tertiärer Vulkanite und ihrer Begleiterscheinungen im Bereich des hessischen Werra-Kaligebietes, Dissertation, Freie Universität Berlin.
- KamLAND Collaboration (2011), "Partial Radiogenic Heat Model for Earth Revealed by Geoneutrino Measurements", Nature Geoscience, 4, 647-651, accessed 8 November 2012, www.nature.com/ngeo/journal/v4/n9/full/ngeo1205.html.
- Knipping, B. (1989), "Basalt Intrusions in Evaporites", Lecture Notes in Earth Sciences, 24, Springer (1989).
- Kpekpasse, A. (2006), Comparison of Fluid Inclusions in Bedded Salt of the Syracuse Formation Versus Salt of the Syracuse Formation in Contact with a Kimberlite Cross-Cutting Dike, MS Thesis, Temple University, Philadelphia, PA.
- Machel, H.G., H.R. Krouse and R. Sassen (1995), "Products and Distinguishing Criteria of Bacterial and Thermochemical Sulfate Reduction", Applied Geochemistry, Vol. 10, pp. 373-389.
- Plant, J.A., et al. (1999), "Uranium Ore Deposits Products of the Radioactive Earth", Reviews in Mineralogy, Vol. 38, pp. 255-319.
- Rempe, N.T. (2011), "Engineered and Natural New Mexico Analogues for Geological Isolation of Heat Generating Waste in Rock Salt", Proceedings of the International High-Level Radioactive Waste Management Conference, Albuquerque, NM, accessed 4 November 2012,
  - $http://cybercemetery.unt.edu/archive/brc/20120623112513/http://brc.gov/sites/default/files/comments/attachments/rempe\_ans\_albuquerque\_.pdf.\\$
- Simmons, A.M. and J.S. Stuckless (2010), "Analogues to Features and Processes of a High-Level Radioactive Waste Repository Proposed for Yucca Mountain, Nevada", USGS Prof. Paper 1779, accessed 5 November 2012, http://pubs.usgs.gov/pp/1779/pdf/PP1779.pdf.

# NEA/RWM/R(2013)10

# Deformation mechanisms and constitutive behaviour during long-term creep of evaporites<sup>1</sup>

Guillaume Desbois, Shiyuan Li, Frank Strozyk, Steffen Abe, Heijn van Gent, Joyce Schmatz, Peter A. Kukla, Janos L. Urai RWTH Aachen University, Germany

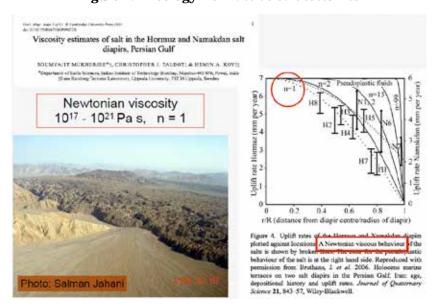


Figure 1: Rheology from active salt tectonics

<sup>1.</sup> The full paper being unavailable at the time of publication, a slightly adapted version of the contribution as presented at the symposium is provided here.

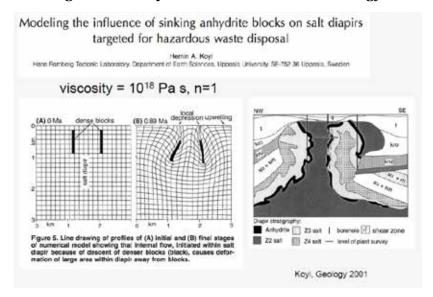


Figure 2: Consequences of Newtonian salt rheology

#### **Deformation mechanisms and rheology**

The constitutive laws governing the deformation of evaporates are closely related to the deformation mechanisms which in turn are a function of microfabric, fluid distribution, strain rate, temperature, etc.

There is a strong controversy in the published literature on this issue.

In the engineering community working on salt mining and repository design, the deformation mechanisms usually concerned are dislocation creep and microcracking.

In the geological community, fluid-assisted grain boundary processes of dissolution-precipitation and recrystallisation are also considered, with associated constitutive laws that predict many orders of magnitude faster creep at low stresses and long time scales.

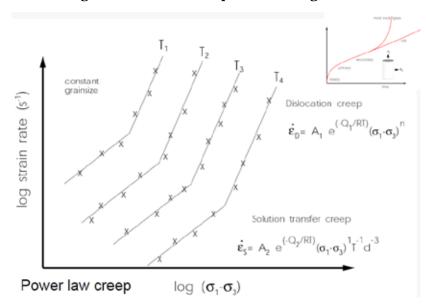


Figure 3: Constitutive equations (Stoffgesetze)

dislocations, subgrains
water assisted dynamic recrystallization

PRESSURE SOLUTION

PLASTICITY, MICROCRACKING

crystal plasticity, microcracking dilatancy, permeability increase

Figure 4: The small scale - material science

Figure 5: Gamma irradiation facility (DFG)

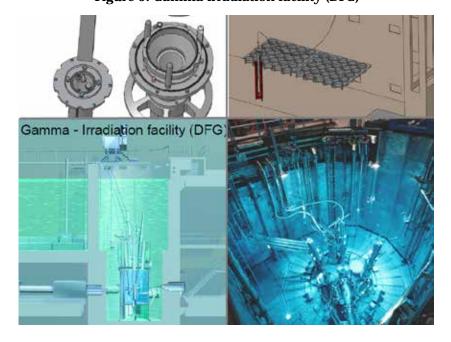
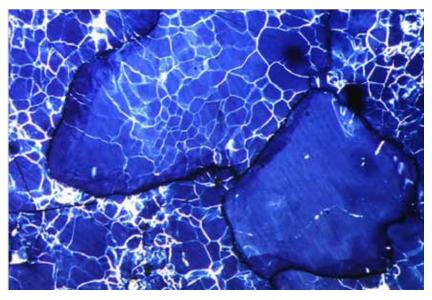
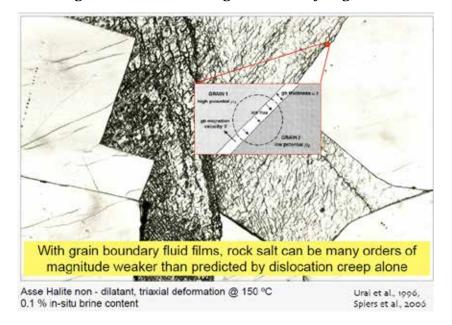


Figure 6: Decorated salt microstructure; weakly deformed Hengelo rock salt



Source: Schieder and Urai, 2006.

Figure 7: Water-assisted grain boundary migration



## Figure 8: Cover of Nature magazine, discussing the topic of weakening rock salt

The majority of Zechstein domal salt underwent extensive dynamic recrystallisation during slow, natural deformation; this process is active because grain boundaries are "active" and contain nm-scale thin fluid films which dramatically increase their mobility (Ural, Spiers, et al., 1986)

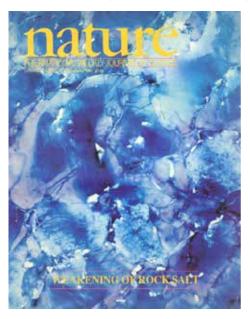
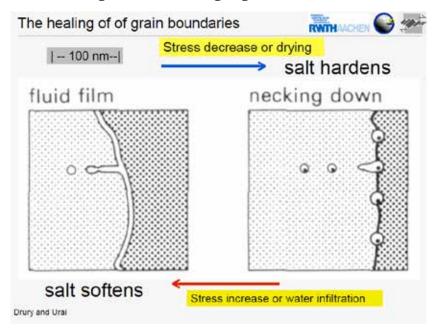


Figure 9: The healing of grain boundaries



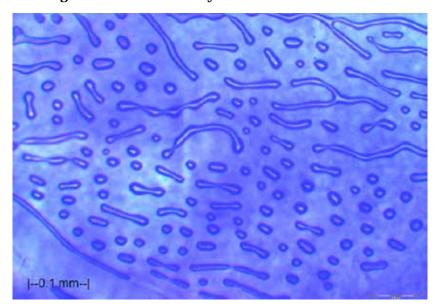
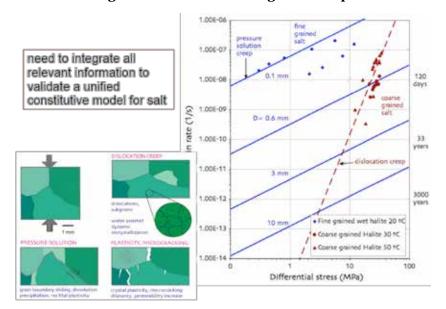


Figure 10: Grain boundary fluid inclusions in rock salt





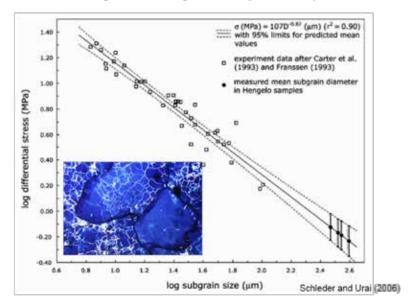
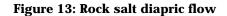
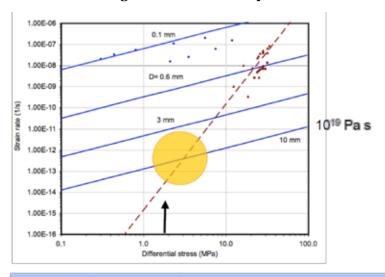


Figure 12: Subgrain size piezometry





In agreement with simultaneous dislocation creep and pressure solution

dislocations, subgrains
water assisted dynamic recrystallization

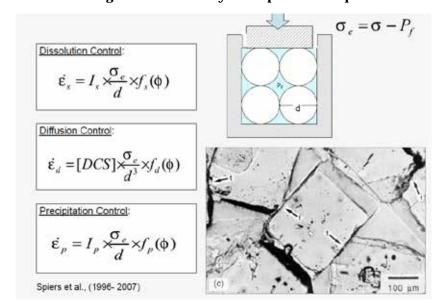
PRESSURE SOLUTION

PLASTICITY, MICROCRACKING

crystal plasticity, microcracking dilatancy, permeability increase

Figure 14: Rock salt deformation mechanisms

Figure 15: IPS theory - compaction creep



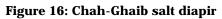
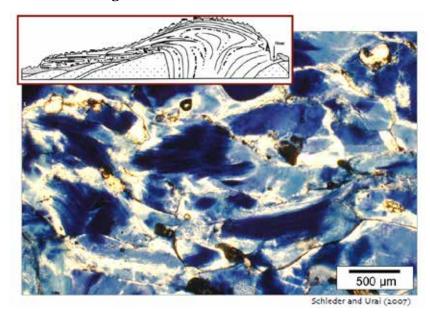




Figure 17: Glacier salt microstructure



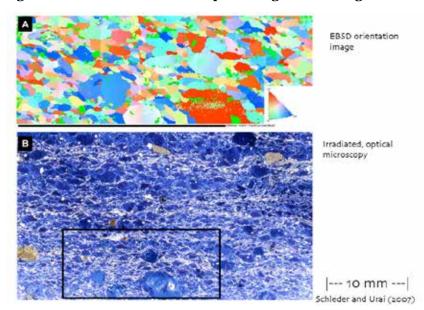
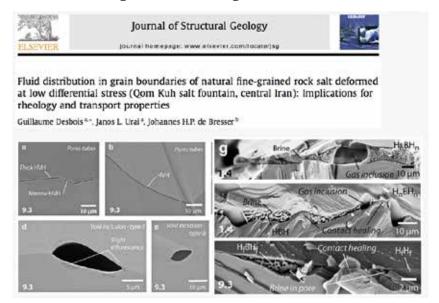


Figure 18: Pressure solution creep in fine-grained, wet glacier salt

Figure 19: An analogue for backfill?



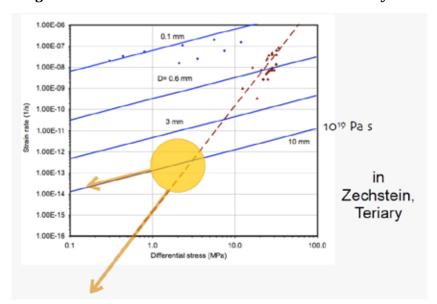


Figure 20: Zechstein - which mechanism is active at very low



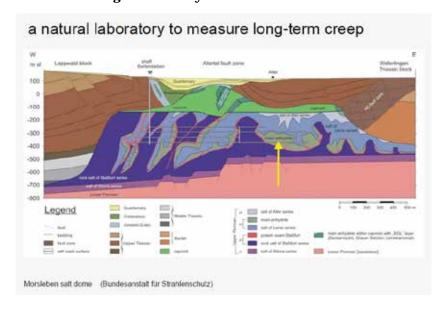
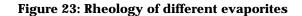
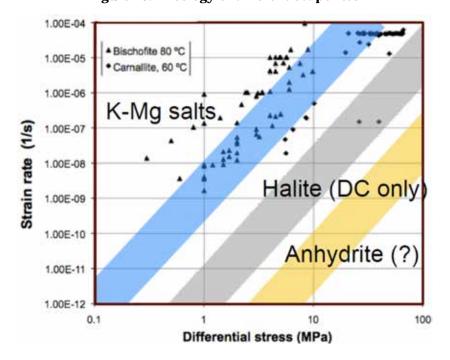




Figure 22: Anhydrite folds and boudins in salt





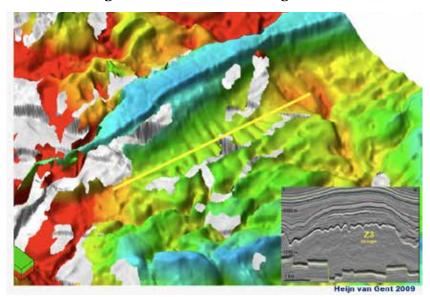


Figure 24: Z3 surface in Gronigen area

Figure 25: Isolated anhydrite stringers in salt

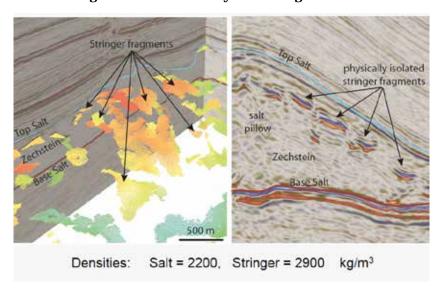
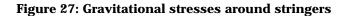
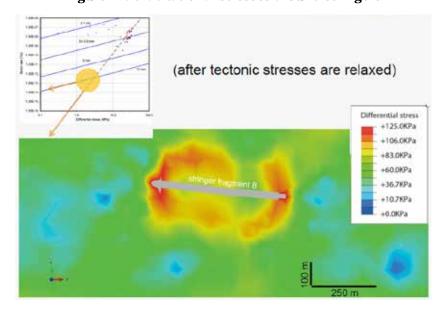


Figure 26: Kinematics of suprasalt sequence





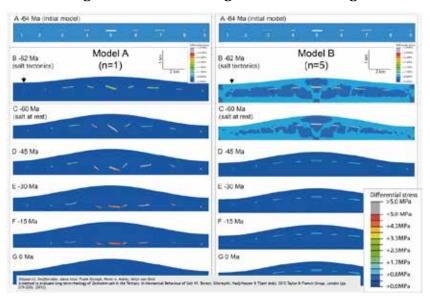


Figure 28: Post-tectonic gravitational sinking

Figure 29: Abstract of an article by S. Li, S. Abe, J.L. Urai

A method to evaluate long-term theology of Zechstein salt in the Tertiary
 Li. S. Abe J.L. Urai

Strictural Geology, Tectorics & Geomechanics, RWTH Aschen University, Lochnesit: 4-20, 52056 Aachen, Germany

F. Strozyk, P.A. Kukla
Geological Institute, RWTH Aachen University, Wullnesit: 2-52056 Aachen, Germany

H. van Gent

Now at 3heff Global Solutions, Resider Park 1, 2288 GS Ryswirk, The Netherlands

All STRACT: The theology of rook talt during slow deformation in nature is controversial. Laboratory experiments and microtractural studies propose that salt can be both Newtonian and non-Newtonian viscous, and deformation of originate of authorities and instingers? substituted in Zechstein salt which has been reconsciously largely macrotre during the Techsity, Coronechanical models amiger flagments in ast is compared with observations of singers in setting data. Essatis show that stranger fragments would stake insadeds on meets through the salt section during a few Mart pressure solution error government. The transpirent of not on terms through the salt section during a few Mart pressure solution reconstruction. The transpirent of solution of meets through the salt section during a few Mart pressure solution reconstruction. The transpirent of control stake insadeds on meets through the salt section during a few Mart pressure solution reconstruction of the formation of the salt state of decreasing by dislocation creep due to very low differential stresses around the stranger studients is understructed that modeling of blocky adaydative stranger staking is an amportant way to obtain the long term theology of the halite, and that the theology of Zechstein salt during the Ternary way dominated by dislocation creep.

Mechanical Bistarten of Salt VE. Benni, Chemychi, Radj Hause & Tipes (selt) 2012 Techs & Freese Geog, London, Lon

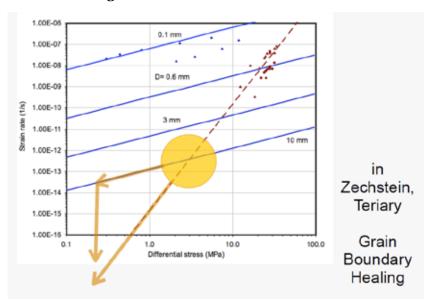
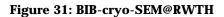
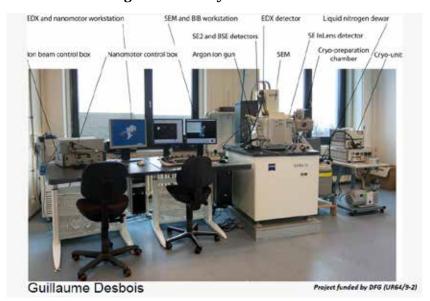


Figure 30: Zechstein salt at low stress





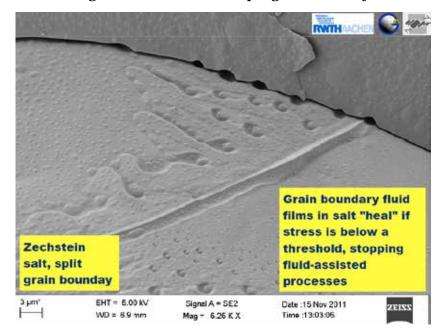
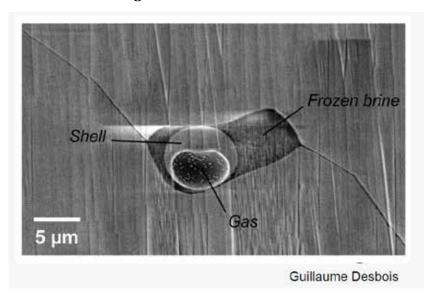


Figure 32: Zechstein salt, split grain boundary

Figure 33: In situ fluids in salt



## **Deformation mechanisms and rheology**

By studying the microstructures and deformation mechanisms in natural evaporite systems, using a novel suite of techniques, we can develop constitutive laws for prediction of the long-term deformation in repositories, based on all the known deformation mechanisms and constrained by natural deformation experiments under conditions not accessible in the laboratory.

These will be consistent with all published data by both communities. More information can be found at www.ged.rwth-aachen.de.

## NEA/RWM/R(2013)10

# Isotope monitoring of water appearances in salt mines: The Polish experience

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Isotope analyses of water appearances in Polish salt mines started in the early 70s of the last century. Soon after it became clear that oxygen and hydrogen stable isotope composition of water creates a unique possibility of determination of the origin of waters in mines and has a crucial role in the risk assessment associated with water inflows. Chemical composition of mine waters, although useful, cannot be used to distinguish between syngenetic (safe) and meteoric (potentially dangerous) water types occurring in mines. The work provides a brief overview of isotope investigations of water appearances in four Polish salt mines (Wapno, Inowrocław and Kłodawa Salt Mines located in northcentral Poland and Wieliczka Salt Mine located in southern Poland) performed over the past forty years.

### Introduction

Polish salt deposits are situated in the north-central and southern part of Poland (Figure 1). They are represented either by salt domes or salt beds, usually covered by water bearing Tertiary and Quaternary formations, sometimes surrounded by older deposits.

There are three groups of water which make their appearance in salt mines: i) meteoric waters of infiltration origin, recharged during both Quaternary and pre-Quaternary climates; ii) syngenetic waters; iii) technological waters and waters of unknown origin. While the first and third category of waters pose a certain danger to the operation of mines, the second category of water is usually safe for salt beds as they occur in a full saturation state and in limited amount, being remnants of the initial salt-forming brines. Sediments surrounding salt deposits are sources of water of meteoric origin which become dangerous when entering the salt formations.

Dissolution of different salt minerals along water flow paths in mines leads to enhanced mineralisation of this water. The type of mineralisation is independent of water origin and is entirely controlled by the chemical composition of the given salt deposit. Therefore, the chemical composition of brines appearing in mines cannot be used as an indicator of the origin of water.

Investigations of brines in Polish salt mines carried out in the past forty years clearly demonstrate that only stable isotope composition ( $^2H/^1H$  and  $^{18}O/^{16}O$  isotope ratios) of water provide valuable information about its origin. To some extent, also the tritium content in water and  $^{14}C$  content in dissolved carbonates can be helpful in determination

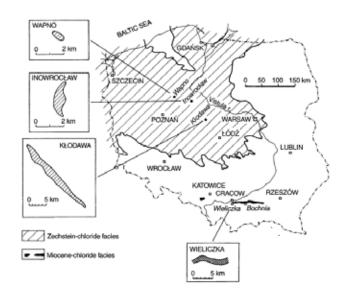


Figure 1: Salinary provinces in Poland, and the investigated areas (Zuber, et al., 1997)

of water origin although they should be treated with a caution at low measured concentrations due to possible isotope exchange with water vapour or carbon dioxide present in the atmospheres of mines. In the following, the Polish experience in determination of water origin in salt mines is briefly described. We focused on four salt mines localised in Wapno, Kłodawa, Inowrocław and Wieliczka (cf. Figure 1).

### Discussion

#### The Wapno Salt Mine

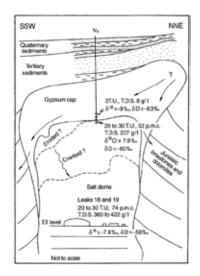
Exploitation in Wapno village started in 1826. Initially it had the form of an open pit gypsum mine. Over the years the open pit mine was transformed into the deep mine. At this time the uppermost Levels I and II were exploited within the gypsum cap of the underlying salt dome. Exploitation of salt started in 1911. In years 1927-1930 two shafts were build (one of them was almost immediately flooded). In 1964 a new shaft started to operate. Initially, the salt was exploited at Levels IV-IX (406-543 m depth). The problem with dangerous water inflows to the mine appeared when the exploitation started at Level III.

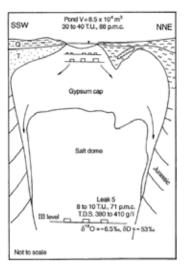
The isotopic characteristics of waters appearing at Level III are presented in Figure 2. The modern origin of waters in the well N1 and at Level III was clearly indicated by the presence of tritium and high <sup>14</sup>C content. Stable isotope composition of waters modified with respect to the values expected for Holocene infiltration suggested the existence of efficient isotope exchange with water of crystallisation of the hydrated salt minerals. It was clear that water appearing at the third level is of meteoric origin. However, this information was ignored by the mine managers. In the night of 4 August 1977, within five hours about 1.5 million cubic meters of water flooded mine corridors and chambers. The catastrophe was probably unavoidable but its large scale and high costs (also including damages of buildings on the surface) were caused by several main reasons: i) intensive exploitation at the third level; ii) excavation of large chambers (15 m wide, 180 m long, up to 12 m high) leading to weakening of safety pillars, iii) too strong explosives used during the exploitation; iv) poor management of the mine ignoring repeated warnings about the infiltration origin of waters appearing at Level III.

Figure 2: Isotopic characteristics of inflows to the Wapno Salt Mine (Zuber, et al., 1997)

Left: north-western part of the salt dome. Right: south-eastern part.

Arrows indicate presumable directions of water flow.





#### The Kłodawa Salt Mine

The Kłodawa deposit is situated in the central part of a 63 km long salt structure. That part pierces through the Mesozoic strata and forms a diapir 26 km long and 0.5-2 km wide. The gypsum-clayey cap is at a depth of 40-500 m, up to 250 m thick, and is surrounded by Quaternary and Tertiary water-bearing sediments. Below the Tertiary sediments, the diapir is surrounded by Mesozoic formations. Salt is exploited at four main levels: 450, 525, 600 and 750 m and several intermediate levels. Three shafts are operated. Yearly production of salt exceeds 600 ktonnes. The volume of exploitation chambers is approximately 18 million cubic meters, with typical dimensions 120  $^{\circ}$  12  $^{\circ}$  12 m.

The isotopic characteristics of water appearances in the Kłodawa Mine are presented in Figure 3. There are three main groups of water identified in the mine on the basis of their isotopic composition. The first group (diamonds in Figure 3) represents syngenetic waters. They are of Zechstein age and represent different stages of evolution of sea water during desiccation of shallow lagoons. They are usually closed within salt pockets and occur in limited amounts; their occurrences usually disappear after some time. Points representing stable isotope composition of these waters form characteristic "hook" shape on the d'H-d¹8O diagram. In some water appearances of measurable amounts of tritium had been found. Its presence in brines has been interpreted as a result of isotope exchange with atmospheric water vapour inside the mine. Their stable isotope composition is shifted towards the WMWL (Figure 3).

The second group of points (squares in Figure 3) represents waters posing a certain risk for the operation of the mine. They are of meteoric origin and of different age. Most of them carry significant evaporation signature and at the present stage of knowledge it is difficult to decide whether the evaporation took place before infiltration or after their appearance in mine passages. Some of these waters were most probably recharged in climatic conditions significantly colder than those prevailing at present in central Poland. Other waters of this type most probably represent Holocene infiltration. In contrast, the appearances no. 7/450 and 14/450 undoubtedly represent pre-Quaternary infiltration, characterised by significantly higher air temperatures than the present ones.

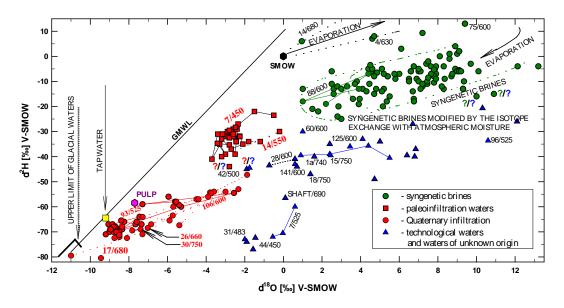


Figure 3: Stable isotope composition of waters appearing in the Klodawa salt mine (selected data)

The third group of points in Figure 3 (triangles) represents technological waters and waters of unknown origin. Technological waters are relatively easy to recognise. Usually they are formed by condensation of atmospheric water vapour or appear as drops on the connectors of pipelines. They create characteristic erosion holes on the floor of mine corridors. Some water appearances within this group are, however, difficult to explain. Their stable isotope composition may result from complicated, multi-stage isotope exchange processes with atmospheric water vapour and/or crystallisation water in salt minerals. This needs further, in-depth investigations.

## The Inowrocław Salt Mine

The Inowrocław salt diapir is about 2.5 km long and 1 km wide. The gypsum cap is at a depth of 6-221 m, while the salt dome is situated at depth of 122-272 m. In the 19<sup>th</sup> century salt was exploited by two underground mines and solution chambers which were created through wells. In 1907 both mines were catastrophically flooded, while the solution chambers were exploited until 1924 to a depth of approximately 180 m. Between 1924 and 1933 a deeper mine was built below the older one, with the first level at a depth of 470 m. Its exploitation began in 1933. Ten exploitation levels have been created since then, each separated by approximately 18 m of salt deposits.

Stable isotope composition of water appearing inside the Inowrocław Salt Mine is presented in Figure 4. The observed pattern is similar to that presented in Figure 3 for the % by points positioned within a characteristic hook trajectory and reflects syngenetic brines. The second group is formed by waters of Holocene infiltration revealing a characteristic shift towards more positive delta values (up to ca. -4 and -40% for d¹8O and d³H, respectively) due to partial evaporation. They pose a serious hazard to mine safety. Technological waters are included in this group. The third group of points in Figure 4 is characterised by d¹8O between ca. -4  $\div$  +2% and d³H from -50  $\div$  -30%. They are located to the right of the GMWL. The origin of these waters is unclear. They can be of meteoric origin, with their isotopic composition modified as a result of isotopic exchange with crystallisation waters of gypsum cap. Those waters may also create a hazard for the mine.

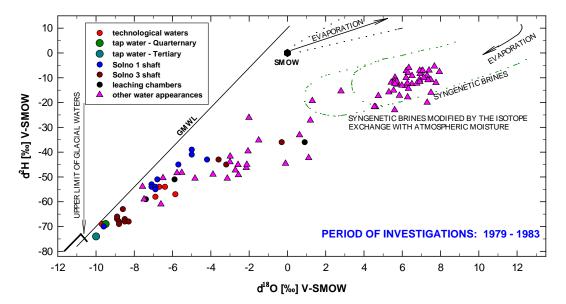


Figure 4: Stable isotope composition of brines in the Inowrocław Salt Mine

Precise interpretation of isotope data from the Inowrocław Salt Mine encounters certain difficulties. The main reasons involve the presence of fresh water which was (and is) used in the exploitation process as well as the presence of strongly evaporated water in some chambers, with the isotopic composition close to that of residual brines. Further difficulties arise from secondary processes which modify the isotopic composition of waters in the mine, such as evaporation, mixing, dissolution of hydrated salts and isotopic exchange with atmospheric water vapour during the migration through cracked salt rocks to the deeper levels of the mine.

#### The Wieliczka Salt Mine

The Wieliczka Salt Mine exploits a Miocene salt deposit. It forms two major structures: i) the lower structure containing strongly tectonically displaced salt; ii) the upper structure consisting of salty clays with large salt blocks. In the south, the salt bed is surrounded by Carpathian flysch deposits. Relatively little is known about the Tertiary water-bearing rocks (Chodenice beds) which are located at the northern boundary of the deposit. During more than seven centuries of uninterrupted exploitation approximately 2 040 chambers with a total volume of 7.5  $^{'}$  106 m³, and over 190 km of galleries in nine levels and several local mid-levels have been created. The Wieliczka salt mine is a world-class monument featuring among the first twelve objects on the UNESCO's World Cultural and Natural Heritage List.

Water discharges to the Wieliczka Salt Mine are among the largest in the Polish salt mines. Up to now about 260 water inflows have been identified. Currently, the total discharge to the mine is approximately 35 m³ per hour, and is dominated by several larger inflows situated at the IV, V and VI levels. Water appearances in the mine have been investigated since the beginning of the 1970s. Their stable isotope composition is presented in Figure 5.

All inflows are of meteoric origin with water ages ranging from the final stages of the last glaciation to modern. Lack of syngenetic waters is a characteristic feature of the Wieliczka Salt Mine. Such waters occur only in certain locations of the deposit, in the form of liquid or liquid/gaseous microinclusions (Dulinski, et al., 1999; Bukowski, et al., 2001). All significant inflows are monitored isotopically. Two of them are particularly interesting.

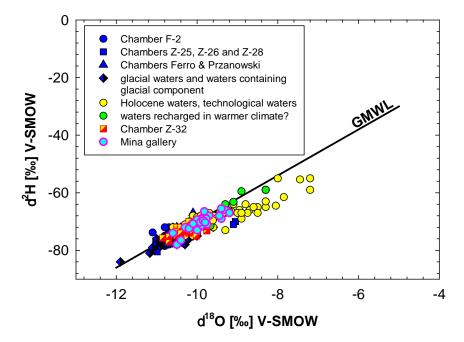


Figure 5: Stable isotope composition of water inflows to Wieliczka Salt Mine

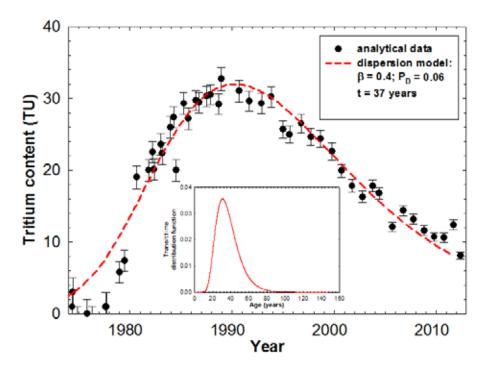
In 1960 an inflow with a discharge rate of 85 m³/day appeared in chamber Z-32 at Level V, as a result of ceiling damage on Level II. Initially, the stable isotope composition of water and lack of tritium suggested inflow of glacial water. Over time, however, the deuterium and ¹8O content has increased, reaching around the year 2000 the delta values typical for Holocene infiltration waters. This evolution was accompanied by a gradual increase of tritium content, reaching a maximum around 1990 followed by gradual reduction of tritium levels, approaching the natural level of tritium in precipitation in the area (Duliński, et al., 2001). The evolution of tritium content in chamber Z-32 is shown in Figure 6. At the present time the discharge rate is at the level of 75 m³/day.

Lumped-parameter modelling (PCFLOW) (Małoszewski and Zuber, 1996) was used to characterise the time scale of water discharging to the chamber. The best fit for the measured tritium levels was obtained assuming a dispersion model of water flow. Parameter b denotes the contribution of tritium-free water, recharged before 1952, i.e. before nuclear tests in the atmosphere. The mean transit time of the tritium-bearing component of flow derived from the modelling was approximately 37 years. The transit time distribution function presented in the insert of Figure 6 suggests significant contribution of flow lines with transit times shorter than ca. 20 years. The tritium data shown in Figure 6 clearly indicate persisting danger to the mine safety stemming the presence of this inflow.

One of the most catastrophic inflows to the Wieliczka Salt Mine occurred in 1992 in the Mina gallery at Level IV. Before World War I this gallery crossed the salt boundary and stopped 20 m beyond the salt deposit within the gypsum and clay formation. Since then the inflow has remained small (1.4-2.9  $\rm m^3/day$ , TDS » 240 g/L) and thought to be safe. Serious troubles began during reconstruction of the gallery in 1992, focusing on closing the inflow. In the night of 13 April 1992, the flow rate suddenly increased to 290-430  $\rm m^3/day$ . Water with high content of suspended matter (ca. 500 kg/m³) was flowing to the gallery. After weeks of recovery work the inflow was stopped and a dam was constructed. Since then water was pumped to the surface to reduce the pressure outside the dam. At the end of 2007 the pumping was stopped.

Figure 6: Record of tritium content in the water inflow to chamber Z-32 in the Wieliczka Salt Mine

Broken line: best fit of lumped-parameter model Insert: transit time distribution function fitted to the measured tritium data



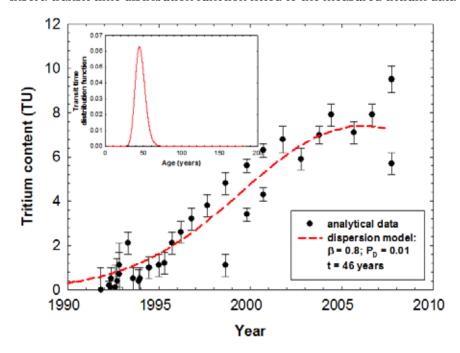
The first isotopic measurements carried out in 1991 pointed out the inflow of glacial water. After the accident, the isotope analyses of inflowing water were performed on a regular basis. Initially, the d²H and d¹8O values scattered widely, suggesting variable conditions and the inflow of water of glacial origin, waters recharged during the Holocene and mixed waters in different proportions. In 1997 the stable isotope composition of water pumped from the Mina gallery stabilised at a level typical for modern infiltration waters in the vicinity of the Wieliczka Salt Mine.

The evolution of tritium content in the inflow to the Mina gallery since 1992 is shown in Figure 7. As in the case of chamber Z-32 data, the measured tritium contents were interpreted in the framework of the lumped-parameter modelling. The best fit of measured and modelled tritium data could be obtained with the aid of the dispersion model. The contribution of the tritium-free component is twice as high as in the case of Z-32 chamber (b = 0.8). The mean transit time of the tritium-bearing flow component is approximately 46 years. The low value of the dispersion parameter (PD = 0.01) and the resulting relatively narrow distribution of transit times (insert in Figure 7) suggest a spatially localised origin of water. After 2007 this inflow was no longer available for analyses.

Water is the most important risk factor in the salt mine industry. Therefore, from the management perspective it is of utmost importance to identify the origin of various water appearances in salt mines and to distinguish between syngenetic (potentially safe) and meteoric (potentially dangerous) water types occurring in this environment.

Figure 7: Tritium content in water inflowing to the Mina gallery and results of numerical modelling

Broken line: best fit of lumped-parameter model Insert: transit time distribution function fitted to the measured tritium data



#### **Conclusions**

The mineralisation of mine waters is largely independent of water origin. Therefore, chemical composition of brines appearing in mines cannot be used as an indicator of the origin of water. Instead, environmental tracers (both stable and radioactive) have a unique capability of providing information about both the origin and the age of mine waters. Long-term experience of using environmental tracers in Polish salt mines shows that stable isotope composition of water (dH and dI8O) is a particularly powerful indicator of the origin of water in the salt mine environment and in most cases allows a clear distinction between syngenetic and meteoric water types. Also, the bomb-tritium is a very useful indicator of the age of water entering the given mine; absence of tritium is a proof that this water was recharged before the bomb era.

The overview presented here of isotope investigations of water appearances in Polish salt mines, performed over the past forty years, clearly demonstrates the value of regular, monitoring-type measurements extending over long periods. Abundant sets of data resulting from such isotope monitoring can then be used with much higher confidence for proper classification of existing and appearing water inflows than is the case for isolated, single measurement campaigns.

#### References

- Bukowski, K., M. Duliński, K. Różański (2001), "Origin of the Badenian Salts from Wieliczka as Indicated by Stable Isotope Composition of Fluid Inclusions", Biuletyn PIG, No. 396, pp. 27-29.
- Duliński, M., et al., (1999), "Stable Isotope Composition of Fluid Inclusions Preserved in Halite Derived from Wieliczka and Bochnia Beds (Southern Poland)", Proc. Int. Symp. on Isotope Techniques in Water Resources Development and Management, Vienna, IAEA-CSP-2/C CD-ROM, International Atomic Energy Agency (IAEA), Vienna, pp. 158-159.
- Duliński, M., et al. (2001), "25-Years of Systematic Measurements of Isotope Composition of Precipitation in Poland", Przeglad Geol., Vol. 49, No. 3, pp. 250-256 (in Polish).
- Małoszewski, P., A. Zuber (1996), "Lumped Parameter Models for Interpretation of Environmental Tracer Data", in Manual on Mathematical Models in Isotope Hydrogeology, IAEA-TECDOC-910, IAEA, Vienna, pp. 9-58.
- Zuber, A., J. Grabczak, A. Garlicki (1997), "Catastrophic and Dangerous Inflows to Salt Mines in Poland as Related to the Origin of Water determined by Isotope Methods", Environmental Geology, Vol. 39, No. 3-4, pp. 299-311.

## NEA/RWM/R(2013)10

# Fluids and fluid migration in salt<sup>1</sup>

## Janos L. Urai, Peter A. Kukla, Guillaume Desbois, Joyce Schmatz, Marc Sadler, Zsolt Schléder, Johannes Schönherr

RWTH Aachen University, Germany

### Fluids in evaporates (solvents and non-solvents)

Fluids can move through evaporites over geologic time, in fractures and in dilatant grain boundary networks. Opening and sealing of transport paths is an important process in this migration. This process can be local or long distance and involve different amounts of fluid. The main controlling parameter in this process is the ratio of fluid pressure and minimum stress.

By understanding fluid migration processes in natural evaporites, we can build quantitative models which describe processes at time scales not accessible in the laboratory, describe the present-day structure of fluids in repository and better predict their response to the changing boundary conditions in repositories.

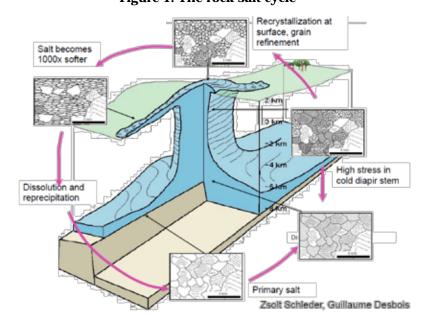
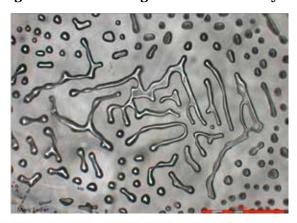


Figure 1: The rock salt cycle

<sup>1.</sup> The full paper being unavailable at the time of publication, a slightly adapted version of the contribution as presented at the symposium is provided here.

Figure 2: Different stages in the rock salt cycle



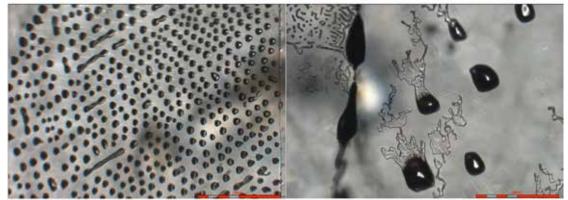
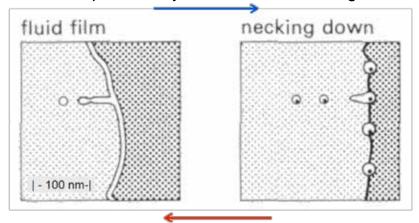


Figure 3: The healing of grain boundaries with water

Compaction, crystallisation, GB healing



Dilatancy, fluid infiltration, recrystallisation

Drury and Ural 1984

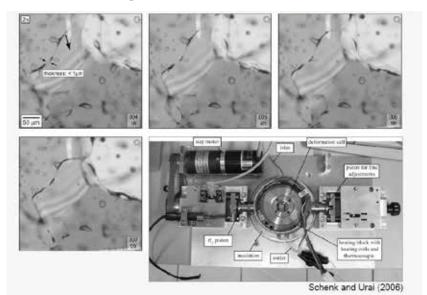


Figure 4: Miniature triaxial cell

Figure 5: A "brine pocket"

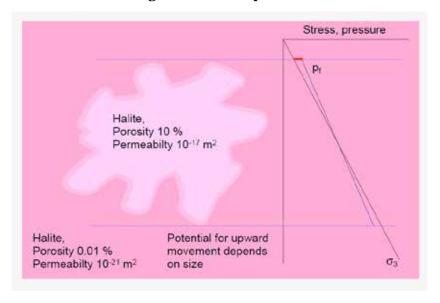
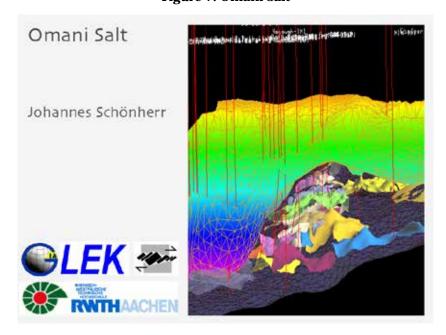


Figure 6: Mobile hydrofactures in gelatin (Paul Bons)

Figure 7: Omani salt



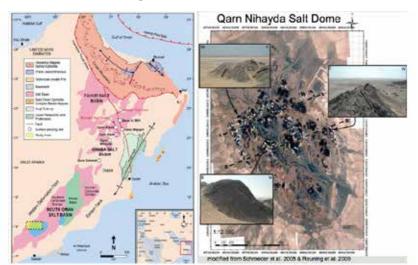
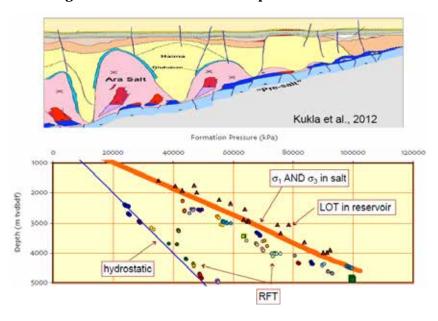


Figure 8: Omani salt basis

Figure 9: In situ stress and fluid pressure in Ara salt



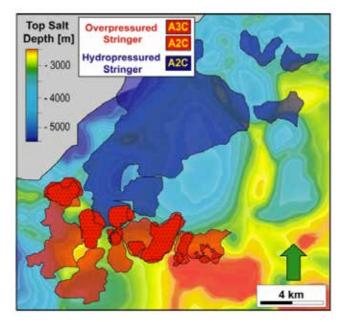


Figure 10: SOSB stringer pressure regimes

Kukla, et al., forthcoming.

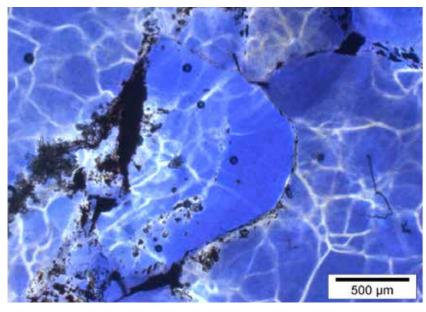


Figure 11: Microstructure of Ara salt

Source: Johannes Schoenherr.

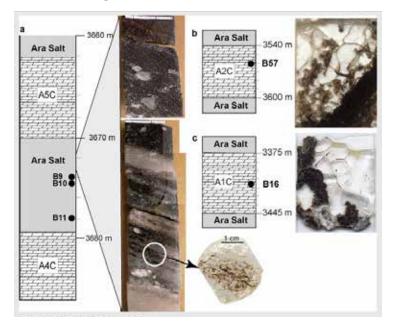
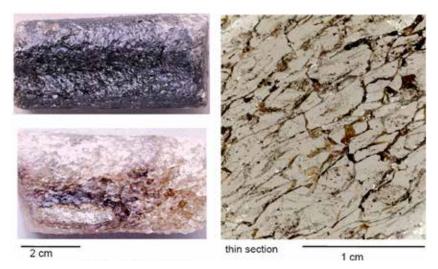


Figure 12: Black salt and bitumen

Source: Schoenherr, et al., 2007, AAPG Bulletin 92.

Figure 13: Core plugs of bitumen-impregnated halite



Source: Schoenherr, et al., 2007, AAPG Bulletin 92.

dilatancy 70 microcracks 1 60 DILATANCY DOMAIN damage 1 Diff. stress (MPa) permeability ↑ 50 creep failure 40 30 compression 20 microcracks ↓ Data from damage ↓ this study 10 permeability ↓ no creep failure 30 50 60 70 80 10 40 Effective stress MPa Dilatancy is only possible at near-zero effective stress - i.e. fluid

Figure 14: Dilatancy boundary for salt

pressure must be lithostatic

Source: Schoenherr, et al., 2007, AAPG Bulletin 92, Popp, et al., 2001.

 $P_{\text{Drine}} \approx \sigma_3. \quad P_{\text{oil}} > \sigma_3 \\ \text{if } P_{\text{oil}} > \sigma_3 + P_c \\ \rightarrow \text{ salt starts to leak} \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable until } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable } P_{\text{oil}} < \sigma_3 \\ \text{salt remains permeable } P_{\text{oil}} < \sigma_3 \\ \text{salt remains } P_{\text{o$ 

Figure 15: Conditions for oil flow through salt

Source: Schoenherr, et al., 2007, AAPG Bulletin 92.

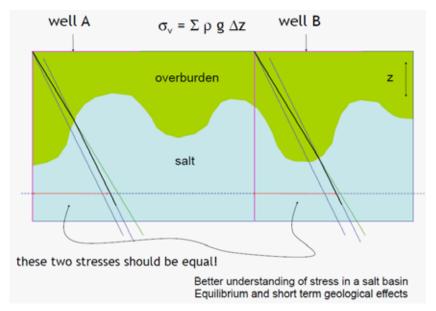
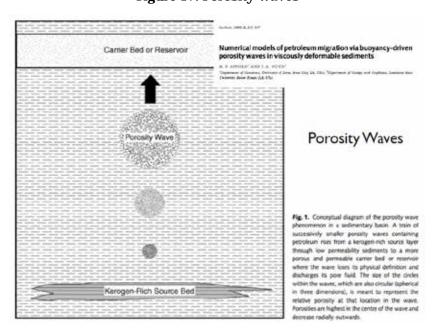


Figure 16: Stress in salt basins

Figure 17: Porosity waves



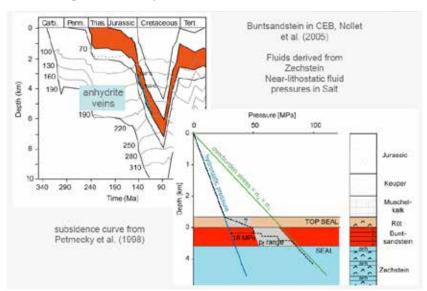
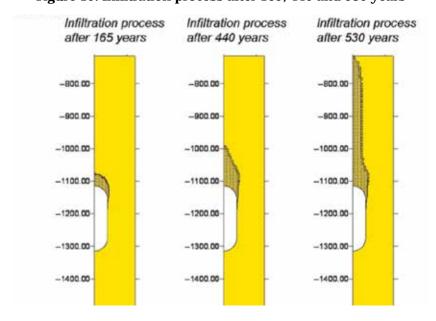


Figure 18: Anhydrite veins from Zechstein-fluids

Figure 19: Infiltration process after 165, 440 and 530 years



Source: Lux, 2009.

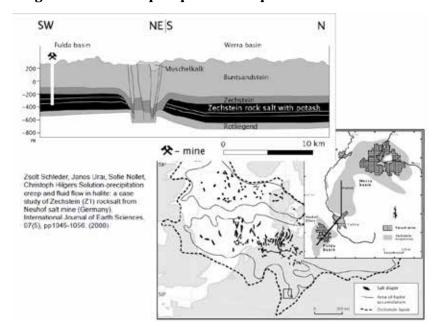


Figure 20: Solution-precipitation creep and fluid flow in halite

Figure 21: The folded halite layers contain halite-filled veins

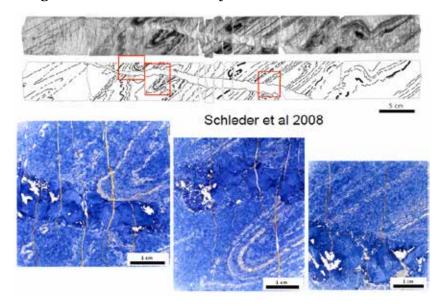
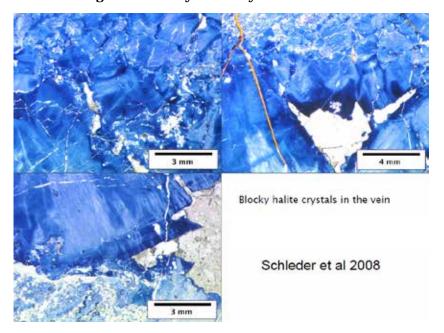




Figure 22: Experimental sealing of fractures in salts





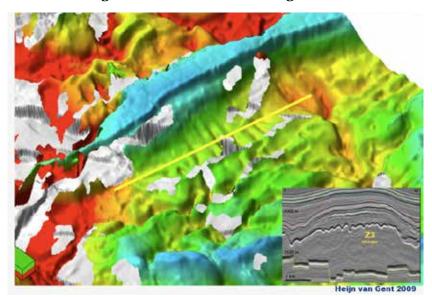
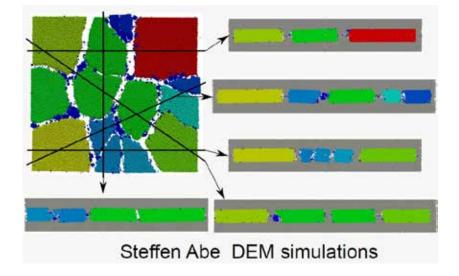


Figure 24: Z3 surface in Groningen area

Figure 25: Rupture process modelled using DEM



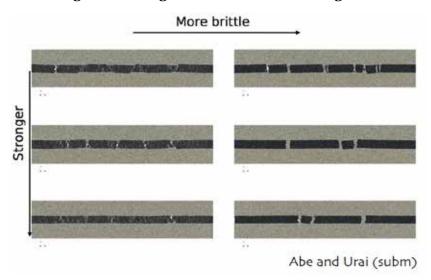


Figure 26: Stringer fractures modelled using DEM

# Deformation and geometry of anhydrite rocks in domal salt structures – results of field studies, mineralogical analyses and thermomechanical experiments

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To evaluate the deformation behaviour of anhydrite rocks during halokinesis and to clarify if these rocks could be migration paths for brines between the salt table and the waste emplacement level, anhydrite rocks of the Allertal zone and Gorleben salt structures, both in Northern Germany, were analysed in detail. The studies on the Hauptanhydrit (z3HA), Gorleben-Bank (z3OSM) and Anhydritmittelsalz (z3AM) are important in the context of long-term performance assessment for HLW repositories in salt domes. Microstructures and mineralogical data were compared to field observations and thermomechanical experiments, showing boudinage and shearing of anhydrite rocks. The naturally deformed anhydrite shows different macro- and microstructural characteristics of the brittle-ductile deformation regime. For anhydrite, solution-precipitation creep and pressure solution are important deformation mechanisms, but further deformation mechanisms (twinning, dislocation creep, subgrain development, bulging and grain boundary migration) are also Microstructural and geochemical analyses noticeable. of matrix halite and of halite inside boudin necks show that rock salt enters the necks by dissolutionprecipitation creep.

### Introduction

The understanding of the deformation behaviour of anhydrite and halite rocks is important for rock salt or potash mining, cavern industries or evaluation of the long-term safety of radioactive waste repositories. The analysis of spatial distribution, structural position and deformational history of anhydrite-bearing rocks is necessary for the prediction of possible migration paths and saline solution reservoirs in salt rocks, prevention of serious problems in drilling and excavating drifts as well as for defining geomechanically instable regions in salt structures (Peach, 1993; Hudec & Jackson, 2007; Hangx, et al., 2011).

In Germany, Permian rock salt (sequence z2, Stassfurt unit) is regarded as a possible host rock for highly active nuclear waste. In salt domes the repository will occupy z2 and z3 (Leine unit), as the emplacement drifts are positioned in z2 and the infrastructure of repository mine is situated mainly in z3. The z3 unit is characterised by stratigraphically and locally changing amounts of anhydrite layers. For long-term safety considerations, the mechanical behaviour of these anhydrite layers and their capacity to store and transmit saline solutions in fissures and faults must receive attention.

To improve the understanding of deformation processes of anhydrite rocks and to evaluate their role as migration paths, thermomechanical experiments, mineralogical analyses and field studies were performed over the last years. In the experiments, composites of single anhydrite layers in halite matrix were deformed under bulk constriction (Zulauf, et al., 2009, 2010), plane strain (Mertineit, 2009; Mertineit, et al., 2012) and oblate strain (Zulauf, et al., 2011) using a thermomechanical deformation rig (a comprehensive summary of experiments is given by Zulauf, et al., 2012). The structures of the experimentally deformed anhydrite layers were analysed and compared to naturally deformed anhydrite beds of the Gorleben salt dome and Allertal zone salt structure (Morsleben), both in Northern Germany. Microstructural and geochemical analyses of halite, situated close to anhydrite and in boudin necks, were carried out to investigate the deformation and healing processes and to constrain the influence of deformation-related, metamorphic brines.

## Methods and samples

In the Gorleben exploration mine and Morsleben mine exposures of anhydrite rocks were studied in detail. Besides observations of deformational features of the Hauptanhydrit in both localities, in the Gorleben salt dome the so-called Gorleben-Bank (z3OSM), an anhydrite layer in the Orangesalz (z3OS) was analysed in more than 180 exposures. The anhydrite bed with a thickness of 3.5 cm to 70 cm (Bäuerle, 2000) is intensively deformed and interrupted. Strong deformation of the Gorleben-Bank is documented by isoclinal folds, boudins and shear bands.

In the Morsleben mine, the Anhydritmittelsalz (z3AM) was investigated in a detailed manner. The Anhydritmittelsalz is characterised by a rhythmic stratification of rock salt and anhydrite layers. The anhydrite layers are up to 2.5 m thick (Behlau, et al., 1997; Behlau & Mingerzahn, 2001). Samples of the anhydrite layers of the Anhydritmittelsalz were collected from the Rüstungskammer 45, shaft Marie, and the Ostquerschlag, both on the second floor level. The layered anhydrite beds are part of Anhydritmittelsalz 3 and 4 (z3AM3/ah and z3AM4/ah, respectively) and are fragmented; on the other hand, Anhydritmittelsalz 5 (z3AM5/ah) with an average thickness of 2 cm, is intensively folded.

Microstructural investigations were carried out using transmitted light microscopy and scanning electron microscopy (SEM). The differential stress in halite close to the rock salt/anhydrite interface, based on the size of halite subgrains was analysed using the following equation:

$$D = 215 * s^{-1.15}$$
 (1)

where D = subgrain size [mm] and s = differential stress [MPa] (Urai, et al., 1987; Franssen, 1994; Schléder & Urai, 2005).

The composite rock salt/anhydrite samples used for the deformation experiments were produced following the method described by Zulauf, et al. (2009). The rock salt was collected from the Asse salt structure (Stassfurt Hauptsalz, z2HS). The anhydrite was collected from the deep borehole Go 1004 in Gorleben (Hauptanhydrit, z3HA).

The anhydrite layers with initial layer thicknesses,  $H_i$ , between 0.85 and 4.5 mm embedded in rock salt, were deformed under bulk plane strain. Further deformation conditions include:  $T = 345\,^{\circ}\text{C}$ ,  $s_{max} = 4.59\,\text{MPa}$ ,  $e_{zmax} = -40\%$ ,  $\dot{e} = 2*10^{-7}\text{s}^{-1}$ . Deformation of halite at laboratory strain rates is only possible at elevated temperature. Each experimental run lasted 2 to 3 weeks. During deformation, the stresses along the Y and Z axis were recorded and compared to the stresses obtained from subgrain analyses. The maximum differential stress  $s_{max}$  of 4.59 MPa was chosen in dependence on natural stresses (e.g. Schléder & Urai, 2005). The deformed samples were analysed using a medical X-ray computer tomograph. Thick and thin sections of undeformed and deformed samples were prepared and analysed.

### **Results**

The anhydrite rocks of both localities, the Morsleben mine and the Gorleben salt dome, are strongly deformed. The deformation structures consist of folds, boudins, fractures, thrusts and shear bands. Fissures and boudin necks are filled with salt, basically halite and carnallite containing high amounts of fluid inclusions.

The documentation of the Gorleben-Bank in the Gorleben salt dome reveals the presence of a secondary mineralisation zone (Zone IV) in 80 of the 180 recorded outcrops of the Gorleben-Bank. However, the thickness of Zone IV in these outcrops varies considerably and averages ~3.2 cm. It only exceeds 10 cm in three outcrops. Polyphase deformation, related to halokinetic processes and stages of the salt dome development (Zirngast, 1991; Bornemann, et al., 2008), is documented by: i) different directions of slickenside in the clayey Zone III of the Gorleben-Bank; ii) segmented, multiple composition of Zone IV (halite and carnallite, often separated by thin clayey or sulphate layers).

The Morsleben repository is located in the Allertal zone salt structure. Two types of folding were determined. In the western part, a shear zone with isoclinals and NE vergent (about  $55^{\circ}$ ) folds is documented. The folds have a large amplitude (up to several hundred metres) and contain the z2 to z4 beds from the salt table to the deepest level of the hinge area. In the eastern part, open, symmetric folds with nearly no vergence and smaller amplitudes are disclosed. The folds contain z4 only at the level of the salt table, z3 only at the level of the repository and z2 only below the mine.

The different styles of folding in the eastern and western part are due to the structure of the rocks beneath the Zechstein salt (Behlau & Mingerzahn, 2001). The Hauptanhydrit behaves brittle during folding and is disrupted. The smaller the angle between the fold limbs, the smaller the resulting Hauptanhydrit blocks. Thus, in the western part of the salt structure, in the shear zone, the fracturing of the Hauptanhydrite is most distinct.

In fold hinges of the Anhydritmittelsalz z3AM5/ah, veil-like opaque material occurs. In these areas granular anhydrite crystals are very small (<50 mm). Cleavage planes are decorated with fluid inclusions rich in halite.

The disclosed boudins (z3AM3/ah and z3AM4/ah), with an average thickness of about 70 cm show different geometries, but symmetric shapes prevail. The boudin block geometry (Goscombe, et al., 2004) can be described as, basically, torn boudins, and rarely drawn boudins.

The grain size at the basal rim is very small (< 50 mm), and high amounts of opaque material are noticeable. Shear bands are more common than folds. The small grain size and the development of the opaque seams suggest pressure solution to be an important deformation mechanism. While the soluble components (halite, anhydrite) were removed, the inert opaque phases remained in place.

Towards the central part of a boudin, fan-shaped, granular and acicular crystals of anhydrite can be distinguished. Often several generations of anhydrite developed. The grain size reaches up to 3.5 mm. In large crystals, undulose extinction, twinning and subgrains are common deformation structures, seldom do fissures prevail. A grain-shape fabric and, close to shear bands, a texture was associated with recrystallisation.

Subgrain analyses of the halite close to anhydrite yield sizes of 59 mm  $\pm$  27 mm to 106 mm  $\pm$  60 mm, corresponding to differential stresses of 3.0  $\pm$  0.7 MPa to 1.9  $\pm$  0.6 MPa. The stress increases with decreasing distance to the anhydrite and reaches a maximum of 3.4  $\pm$  1.4 MPa at the halite/anhydrite interface (subgrain size = 52 mm  $\pm$  25 mm).

Experimental results show similar structures as observed in Anhydritmittelsalz or Gorleben-Bank. In cases where the anhydrite layer was orientated perpendicular to the principal stretching axis, the anhydrite layer shows Mohr-Coulomb fractures. The walls

along these fractures were thrust on top of each other, and the space between the hanging and foot wall is filled with salt. Folding of the layer did not occur, but the layers are bent and show tension gashes. The layer thickness did not change during deformation.

In cases where the anhydrite layer was orientated perpendicular to the principal shortening axis, torn boudins developed by extensional fractures. The number of boudins and their average width ( $W_a$ ) strongly depend on the initial layer thickness  $H_i$  and can be described by following equation:

$$W_a = -0.2 + 1.4 * H_i$$
 (2)

The boudin aspect ratio ( $W_d = W_a/H_i$ ) is calculated at ~1.4. A similar relation between layer thickness and wavelength (width) of boudins has been found for bulk constriction and bulk flattening (Zulauf, et al., 2012). The boudin shape in general is largely rectangular to triangular, but irregular shapes are also present. The boudin block geometry (Goscombe, et al., 2004) can be described as symmetric torn boudins. The boudins are generally rotated, especially in the middle of the sample due to maximum flow of the halite matrix.

In the halite, subgrain sizes of 43 mm  $\pm$  12 mm to 58 mm  $\pm$  21 mm result in a differential stress of 3.6  $\pm$  0.7 MPa. Peak stresses close to the anhydrite layer, grain boundaries and impurities, however, reach values of 7.3  $\pm$  1.5 MPa (subgrain size = 22  $\pm$  7 mm). These stresses are compatible with the stresses recorded by the load cells of the machine during the experiments.

### **Discussion**

All natural and experimental macro- and microstructures suggest the investigated anhydrite rocks to have been deformed in the brittle-ductile deformation regime, which is compatible with literature data (Müller, et al., 1981; Dell'Angelo & Olgaard, 1995; Zulauf, et al., 2010).

At the basal rim of the boudins, the interaction between the halite matrix and anhydrite is most pronounced. The deformation structures are thus striking here; almost no sedimentary structures or relicts of former anhydrite generations are left. The major deformation mechanisms are solution-precipitation creep and pressure solution, as is shown by seams and veils of opaque and insoluble material, e.g. magnesite. Further evidence for fluid activity are high amounts of fluid inclusions in the surrounding halite, water-bearing mineral phases like polyhalite, and the low bromide content in halite of boudin necks (Mertineit, et al., 2012).

Towards the central parts of the boudins, the deformation mechanism of anhydrite changes. Evidence for pressure solution (e.g. stylolite) is still present, but intracrystalline deformation prevails. Typical deformation microstructures are undulose extinction, subgrains, bends and kinks of long and/or prismatic crystals, and deformation twins. In certain cases bulging and grain boundary migration were observed. Fracturing of large anhydrite crystals and of the entire rock is disclosed. This is evidence for a second deformation event under different deformation conditions. In high-strain domains, the anhydrite is pervasively recrystallised and shows a shape-preferred orientation and a texture (Passchier & Trouw, 2005). To determine the kind of texture and the active deformation mechanisms, further investigations on the microstructures, e.g. electron backscatter diffraction analyses (EBSD), are necessary (Heidelbach, et al., 2001; De Paola, et al., 2008; Hildyard, et al., 2009).

Most variations in the thickness of the Gorleben-Bank are due to deformation processes like folding, shearing and boudinage, occurring during the halokinetic uprise of the salt. Primary, by sedimentation or diagenesis originated variations in the layer thickness play a minor role. For the Gorleben salt dome, halokinetic processes involving intensive deformation were active several times. During the Röt to Muschelkalk time, the pillow stage of the salt dome evolution started (Zirngast, 1991) and reached its maximum

during Keuper to Dogger. The diapiric development, including penetration of the overburden, lasted from Malm to Lower Cretateous and continued in the Upper Cretateous and Tertiary periods.

In our experiments the anhydrite was deformed in the brittle-ductile regime, characterised by twinning, kinking and fracturing (Mertineit, 2009). Folding did not occur probably due to the lack in confining pressure and/or the mechanical anisotropies in the undeformed anhydrite (Zulauf, et al., 2009, 2010, 2011; Mertineit, 2009; Mertineit, et al., 2012).

Compared to field observations, several differences, but also similarities are noticeable. The experimentally deformed anhydrite behaved in the brittle-ductile manner, whereas brittle behaviour persists. Brittle shear planes (Mohr-Coulomb fractures) developed instead of folds. Crystal plastic deformation, documented by bent crystals, was active, but did not play a major role, whereas brittle deformation in the form of microfracturing predominates. The torn geometry of the experimentally produced boudins is compatible with brittle tensile fracturing.

The anhydrite exposed in the salt mines, was also deformed in the brittle-ductile regime. Similar structures like those produced experimentally (torn boudins, brittle shear planes, and fractures) are noticeable. But in naturally deformed anhydrite additional structures are present. These differences could be related to differences in the external deformation conditions (strain rate, confining pressure, temperature, geochemical environment, fluid activity) and to differences in composition of naturally and experimentally deformed anhydrite.

The strain rate  $\dot{e}$  in the experiments was set at  $2^*10^{-7}s^{-1}$ , strain rates in nature are between  $10^{-16}$  and  $10^{-8}s^{-1}$  (Jackson & Talbot, 1986). Thus the temperature for the experiments was chosen to be  $345^{\circ}$ C. The experiments were carried out in an open system, meaning that during deformation released gases and fluids could disappear. Another difference is the lack of confining pressure, which could explain the lack of folds. Despite these shortcomings, the experiments produce important and basic data and are a helpful tool to understand and quantify geological processes.

## **Conclusions**

Halokinetic salt tectonics cause complex deformation structures in anhydrite rocks. A characteristic decrease of the number of anhydrite blocks is documented as well as increasing distances between anhydrite blocks in higher parts of salt diapirs. The size of anhydrite blocks or boudins depends mainly on fold geometry [wideness of folds; see e.g. small blocks of Hauptanhydrit on higher horizons at the western flank of Allertal zone salt structure (Morsleben) with closely isoclinal folds], as well as thickness of anhydrite rocks, spatial orientation and magnitude of stresses, especially in case of shearing.

The 3-D model of the Gorleben salt dome shows the complex internal structure and the mostly isolated position of anhydrite blocks. This conclusion is based on extensive data from exploration drillings, mapping and geophysical measurements (ground penetrating radar – GPR). The use of the 3-D model allows a distinctly better prediction of the distribution of anhydrite blocks in the salt structure and thus, a strongly better localisation of suitable areas for disposal of radioactive waste.

Most of the studied anhydrite samples collected from the Morsleben mine and Gorleben salt dome consist of several strongly deformed anhydrite types and generations. The presence of various deformation mechanisms (pressure solution, crystal plasticity and brittle fracture) suggest the anhydrite was deformed in the brittle-ductile regime. The number of boudins and their average size depends strongly on the initial layer thickness. Geochemical and microstructural data of halite sampled in boudin necks suggest that this halite results from reprecipitation of previously dissolved halite inside an open fracture.

## References

- Bäuerle, G. (2000), Geochemisch-mineralogische Untersuchungen zur Genese, Lösungs- und Gasführung der Gorleben-Bank (Zechstein 3) des Salzstocks Gorleben, Dissertation, Technische Universität Clausthal, 147 pp. (unpublished).
- Behlau, J., G. Mingerzahn and O. Bornemann (1997), ERA Morsleben. Erarbeitung eines geologischen Lagerstättenmodells Morsleben, Bundesanstalt für Geowissenschaften und Rohstoffe, Abschlußbericht, AP 9M 2124601100, Archiv.-Nr. 116872, 73 pp.
- Behlau, J. and G. Mingerzahn (2001), "Geological and Tectonic Investigations in the Former Morsleben Salt Mine (Germany) as a Basis for the Safety Assessment of a Radioactive Waste Repository", Engineering Geology, 61, 83-97.
- Bornemann, O., et al. (2008), "Description of the Gorleben Site Part 3: Results of the Geological Surface and Underground Exploration of the Salt Formation", Geologisches Jahrbuch, Reihe C, Heft 73, E. Schweizerbart'sche Verlagsbuchhandlung, Stuttgart.
- Dell'Angelo, L.N. and D.L. Olgaard (1995), "Experimental Deformation of Fine-Grained Anhydrite: Evidence for Dislocation and Diffusion Creep", J. Geophys. Res., 100 (B8), 15425-15440.
- De Paola, N., et al. (2008), "Fault Zone Architecture and Deformation Processes Within Evaporitic Rocks in the Upper Crust", Tectonics, 27, TC4017, 21 pgs., doi: 10.1029/2007TC002230.
- Franssen, R.C.M.W. (1994), "The Rheology of Synthetic Rock Salt in Uniaxial Compression", Tectonophysics, 233, 1-40.
- Goscombe, B.D., C.W. Passchier and M. Hand (2004), "Boudinage Classification: End-Member Boudin Types and Modified Boudin Structures", J. Struct. Geol., 26, 739-763.
- Hangx, S.J.T., C.J. Spiers and C.J. Peach (2011), The Mechanical Behavior of Anhydrite and the Effect of Deformation on Permeability Development Implications for Caprock Integrity During Geological Storage of CO<sub>2</sub>", Energy Procedia, 4, 5358-5363.
- Heidelbach, F., I.C. Stretton and K. Kunze (2001), "Texture Development of Polycrystalline Anhydrite Experimentally Deformed in Torsion", Int. J. Earth Sciences, 90, 118-126.
- Hildyard, R.C., et al. (2009), "Microstructural Analysis of Anhydrite Rocks from the Triassic Evaporites, Umbria-Marche Apennines, Central Italy: An Insight into Deformation Mechanisms and Possible Slip Systems", J. Struct. Geol., 31, 92-103.
- Hudec, M.R. and M.P.A. Jackson (2007), "Terra Infirma: Understanding Salt Tectonics", Earth Science Reviews, 82, 1-28.
- Jackson, M.P.A. and J.C. Talbot (1986), "External Shapes, Strain Rates, and Dynamics of Salt Structures", Geological Society of America, Bulletin 97/3, 305-323.
- Mertineit, M. (2009), Faltung und Boudinage einer Anhydritlage in Halitmatrix unter ebenen Verformungsbedingungen: Ergebnisse thermomechanischer Experimente, Thesis, Goethe University, Frankfurt/Main, 77 pp. (unpublished).

- Mertineit, M., et al. (2012), "Mechanical Behavior of Anhydrite Rocks: Results of Field Investigations, Mineralogical-Geochemical Studies and Thermomechanical Experiments", in P. Bérest, et al. (Eds.), Mechanical Behavior of Salt VII: Proceedings of the 7th Conference, Taylor & Francis Group, London, pp. 123-129.
- Müller, W.H., S.M. Schmidt and U. Briegel (1981), "Deformation Experiments on Anhydrite Rocks of Different Grain Sizes: Rheology and Microfabric", Tectonophysics, 78, 527-543.
- Passchier, C.W. and R.A.J. Trouw (2005), Microtectonics, Springer-Verlag, Berlin, Heidelberg, 366 pp.
- Peach, C.J. (1993), "Deformation, Dilatancy and Permeability Development in Halite/Anhydrite Composites", in M. Ghoreychi, et al. (Eds.), The Mechanical Behaviour of Salt, Proceedings of the 3<sup>rd</sup> Conference, Paris, Trans Tech Publications, pp. 53-166.
- Schléder, Z. and J.L. Urai (2005), "Microstructual Evolution of Deformation-Modified Primary Halite from the Middle Triassic Röt Formation at Hengelo, The Netherlands", Int. J. Earth Sci., 94, 941-955.
- Urai, J.L., et al. (1987), "Deformation Mechanisms Operating in Naturally Deformed Halite Rocks as Deducted from Microstructural Investigations", Geol. Mijnbouw, 66, 165-176.
- Zirngast, M. (1991), "Die Entwicklungsgeschichte des Salzstocks Gorleben Ergebnis einer strukturgeologischen Bearbeitung", *Geol. Jb.*, A 132, Hannover.
- Zulauf, G., et al. (2009), "Experimental Deformation of a Single-Layer Anhydrite in Halite Matrix Under Bulk Constriction Part 1: Geometric and Kinematic Aspects", J. Struct. Geol., 31, 460-474.
- Zulauf, G., et al. (2010), "Experimental Deformation of a Single-Layer Anhydrite in Halite Matrix Under Bulk Constriction, Part 2: Microfabrics and Deformation Mechanisms", J. Struct. Geol., 32, 264-277, doi: 10.10.16/j.jsg.2009.12.001.
- Zulauf, J., et al. (2011), "Tablet Boudinage of an Anhydrite Layer in Rock-Salt Matrix: Results from Thermomechanical Experiments", J. Struct. Geol., 33, 1801-1815, doi: 10.1016/j.sg.2011.09.006.
- Zulauf, G., et al. (2012), "Boudinage of Anhydrite in Rock-Salt Matrix: The Impact of Bulk Finite Strain Geometry", in P. Bérest, et al. (Eds.), Mechanical Behavior of Salt, Proceedings of the 7<sup>th</sup> Conference, Paris, Taylor & Francis Group, London, pp. 65-70, ISBN 978-0-415-62122-9.

## **Annex**

**Figure 1: Introduction** 

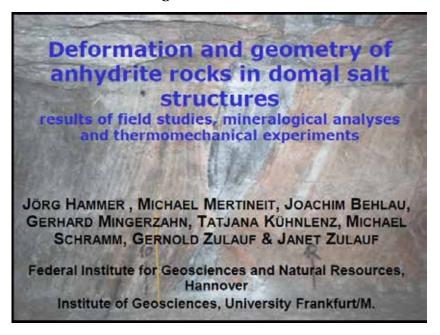


Figure 2: "Hauptanhydrite scenario"

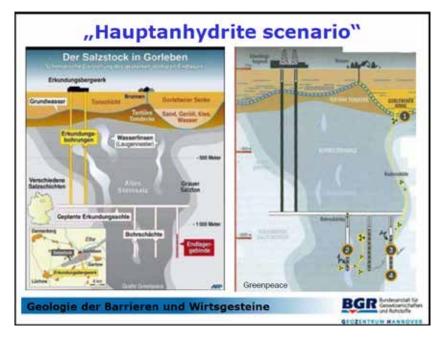


Figure 3: Anhydrite studies are necessary

## Anhydrite studies are necessary

- Safety assessment of a potential HLW repository in salt formations requires evidences for the integrity of the geological barrier.
- The lack of continuous water pathways from salt table to waste emplacement levels in repository has to be shown.
- The aim of our salt geological, mineralogical and experimental studies is to show, that there arn't pathways through anhydrite or at interfaces between rocksalt and more competent anhydrite layers from salt table to waste emplacement level

Geologie der Barrieren und Wirtsgesteine



Figure 4: Outline of the presentation

## Outline of the presentation

- Observations during exploration of Gorleben salt dome (Hauptanhydrite, z3HA; Gorleben-Bank, z3OSM)
- Studies of anhydrite rocks in Morsleben mine (studies of Anhydritmittelsalz; z3AM)
- Thermomechanical experiments deformation of anhydrite layers)

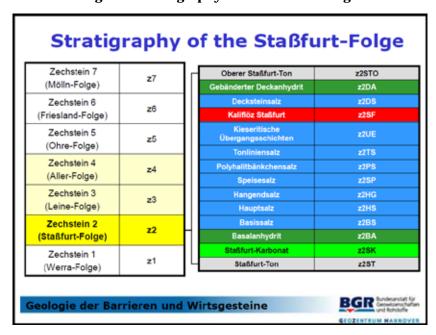
Aim: better understanding of deformation processes for long-term safety considerations



Figure 5: Boudinage and shearing of Hauptanhydrite/ Anhydritmittelsalz/Gorleben-Bank in Gorleben salt dome



Figure 6: Stratigraphy of the Staßfurt-Folge



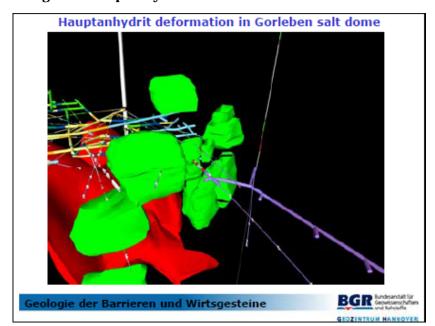
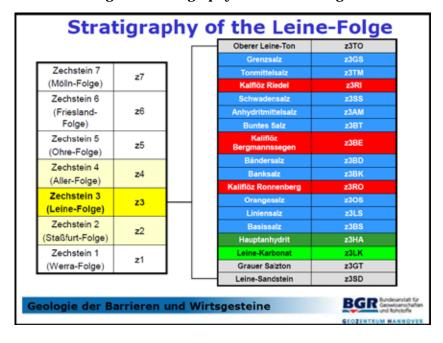


Figure 7: Hauptanhydrit deformation in Gorleben salt dome

Figure 8: Stratigraphy of the Leine-Folge



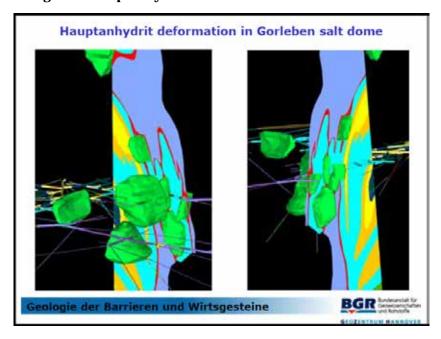
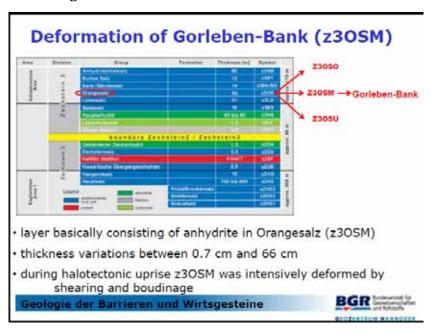


Figure 9: Hauptanhydrit deformation in Gorleben salt dome

Figure 10: Deformation of Gorleben-Bank (z3OSM)



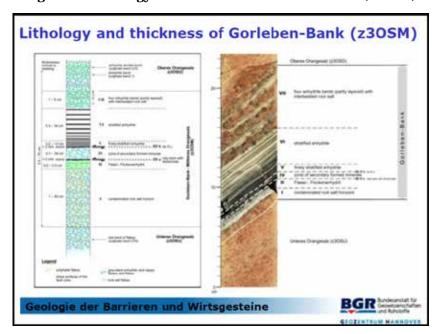
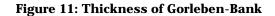
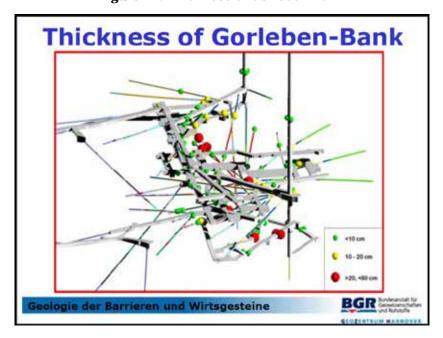


Figure 11: Lithology and thickness of Gorleben-Bank (z3OSM)





Thickness

0,8 cm

23,7 cm

Figure 12: Thickness

Figure 13: Secondary mineralisation (Zone IV)





Figure 14: Deformation/boudinage of Gorleben-Bank







Figure 16: Fractures in Gorleben-Bank

Figure 17: Deformation and healing of Gorleben-Bank





Figure 18: Deformation of Gorleben-Bank (Bohrort 1.8)



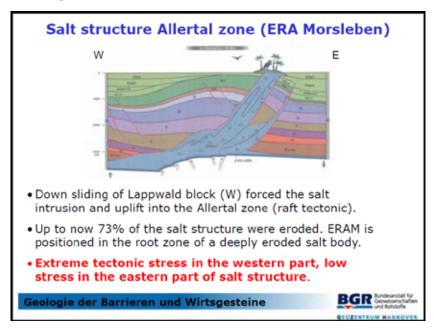




Figure 20: Shear folding, Marie shaft (contact z3LS/z2SF)

Figure 21: Shear folding, Marie shaft





Figure 22: Vergent folding, Marie shaft

Figure 23: Vergent folding, Marie shaft



Figure 24: Boudinaged and folded anhydrite layers of the Anhydritmittelsalz (z3AM), -231 m level, Marie shaft, Morsleben



Figure 25: Sampling of z3AM in Morsleben

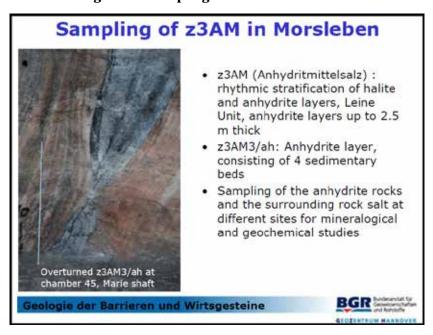
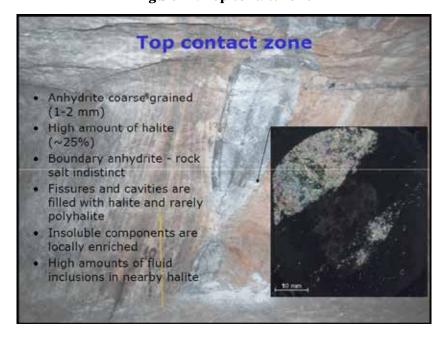




Figure 26: Basal contact zone

Figure 27: Top contact zone



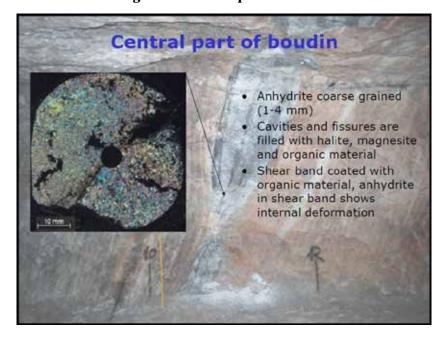
Several generations and types of anhydrite, affected by pressure solution (left)

• Fluid inclusions in halite close to the anhydrite layer (below)

I Polarizers

Figure 28: Microstructures at the anhydrite-halite boundary

Figure 29: Central part of boudin



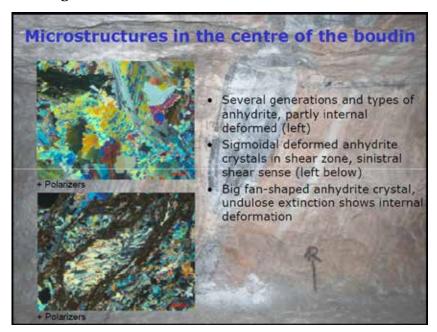
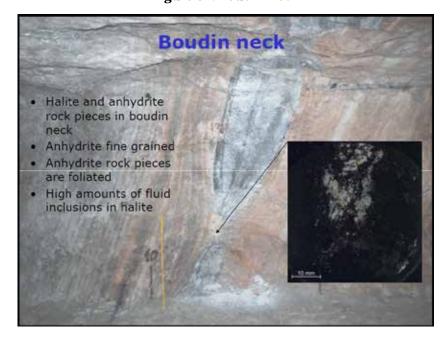


Figure 30: Microstructures in the centre of the boudin





Microstructures in boudin neck

- Few crystals are deformed. Clear crystals without impurities developed later or postkinematically
- Fragments of foliated anhydrite fock
- Fluid inclusions in halite

Figure 32: Microstructures in boudin neck

Figure 33: Conclusions z3AM studies - halite

## Conclusions z3AM studies - halite

- · Halite was deformed in the viscous deformation regime.
- Dissolution-reprecipitation of halite, dislocation creep and subgrain formation/rotation are major deformation mechanisms of halite.
- Halite contains locally high amounts of fluid inclusions, specially in the boudin neck. There are several generations and types of inclusions (bubbles, channels, fluid films on grain boundaries, intracrystalline inclusions), usually in different orientations.



Figure 34: Conclusions z3AM studies - anhydrite

# Conclusions z3AM studies - anhydrite

- The anhydrite was deformed in the brittle-viscous deformation regime.
- Pressure solution and solution-precipitation creep seem to be important deformation mechanism, although evidences for crystalplastic deformation (dislocation creep, subgrain development, grain boundary migration) are present.
- Fractures are filled with salt and, in shear bands, with insoluble components (magnesite, organic material) and deformed fragments of host rock.



Figure 35: Experimental studies - material properties

	halite	anhydrite
composition	NaCl	CaSO <sub>4</sub>
density (g/cm <sup>3</sup> )	2.1-2.2	2.9 - 3.0
hardness (Mohs)	2	3 – 3.5
viscosity η [Pa*s](1)	3*1013 (2)	8*1014(3)
viscosity contrast m	~27	
stress exponent n (1)	5.7 (2)	5 (3)
deformation mechanisms	climb-controlled dislocation creep (rotation recrystallization) and strain hardening	twinning, kinking, microfracturing (brittle-viscous)

Further deformation conditions

Initial sample dimensions:
60\*43\*59 mm (X\*Y\*Z)

Initial layer thickness H;
0.85 - 4.5 mm +/- 0.02 mm

T = 345°C

Figure 36: Further deformation conditions

Figure 37: Deformation geometry

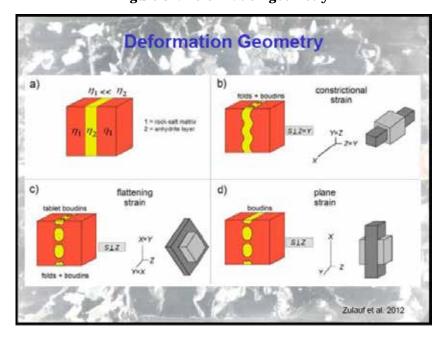


Figure 38: CT image of deformed anhydrite layer (constructional deformation, halite matrix not shown)

Left - oblique view, right - top view

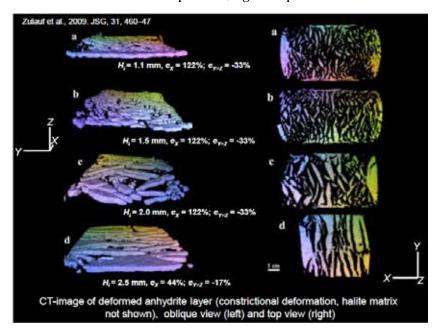
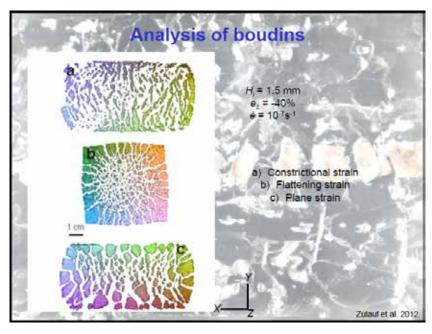


Figure 39: Analysis of boudins



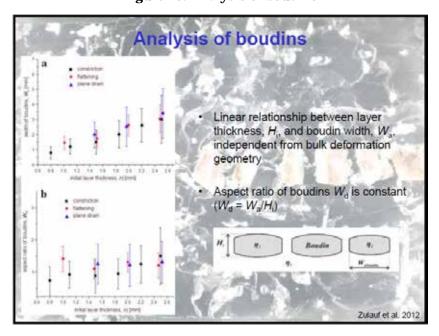


Figure 40: Analysis of boudins

Figure 41: Comparison of natural observations and experiments

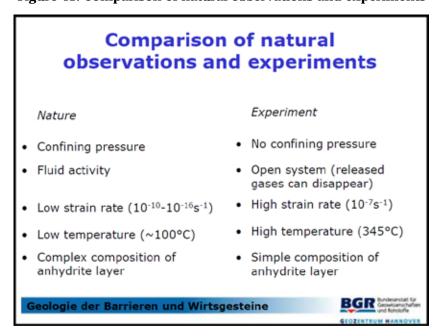


Figure 42: Comparison of natural observations and experiments

# Comparison of natural observations and experiments

### Nature

- Various boudin geometries like torn, drawn,..., boudins
- Dissolution-reprecipitation of halite, dislocation creep and subgrain formation are major deformation mechanisms of halite
- Brittle-ductile behavior of anhydrite, fracturing, crystal plastic behavior, pressure solution

## Experiment

- · Torn boudins, rotated
- Dislocation creep and subgrain formation are major deformation mechanisms of halite
- Brittle-ductile behavior of anhydrite (bent crystals, serrated grain boundaries), but microfracturing predominates



## NEA/RWM/R(2013)10

## Integrity of rock salt formations under static and dynamic impact

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Natural analogues from potash and rock salt mining are unique opportunities to support reasoning and validation of geomechanical modelling as essential tools to prove the integrity of saliferous barriers. Salt formations are naturally gas- and water-tight but can lose their geomechanical integrity and leak tightness under two circumstances: i) if the groundwater pressure or a gas pressure becomes higher than the acting minimal stress in the salt formation; ii) under dilatant conditions. The first process seems to be the most relevant process for the overall barrier integrity. The stress may be lowered due to the acting extensional conditions either by convergence processes around a cavity or by thermo-mechanically-induced elevations of the structure. Based on practical examples, which are classified in geological and industrial analogues, the geomechanical conditions resulting in a loss of tightness will be discussed and supported by means of geomechanical computation models.

### Introduction

Salt deposits have been used for the extraction of mineral resources for millennia. In the  $20^{\text{th}}$  century new fields of application have been added, such as the storage of hydrocarbons in caverns and the disposal of toxic and radioactive waste in domal and bedded salt formations. The disposal of chemo-toxic and radioactive waste in salt rock benefits mainly from the tightness of the salt rocks ensuring the principle of complete enclosure of the waste away from the biosphere.

Noseck, et al. (2008) investigated natural analogues as supplementary reasoning to mathematically performed long-term safety studies with evidence for repository with heat-generating waste in rock salt. The authors distinguish between industrial (< 150 years), archaeological (150...10000 years) and geological (> 10000 years) analogues.

Potash mining with its over 150 year history starting from the Stassfurt region in Germany (Bergmannsverein, 2002) offers unique insights into salt deposits. Due to the extensive insights in mined underground openings, numerous examples of natural analogues exist for the investigation and demonstration of the isolation potential of salt and its integrity behaviour under geogenic and anthropogenic stresses belonging to the industrial and geological categories. These practical experiences are of distinguished relevancy for the heat-generating radioactive waste in salt and have to be pursued regarding their impact for understanding the geomechanical long-term processes. As shown in examples below, it becomes possible to demonstrate the integrity and tightness of saliferous barriers for the required period of time for a long-term repository in salt formations by using geological analogues.

Due to the disposal of heat-generating waste additional thermo-mechanical stresses are induced in the surrounding geological barriers with elevations on the earth's surface in the meter range (Eickemeier, et al., 2012). Subsidence processes over potash and rock salt mines may result in the same order of deformation. However, with respect to the understanding of the fundamental processes of barrier integrity, it can be stated that

from a mechanical point of view, there is no difference regarding the loading of the barrier if uplift or subsidence processes are investigated.

Aiming on the disposal of both chemo-toxic and heat-generating radioactive waste in a salt deposit, the processes which are most relevant for the barrier integrity of salt are acting in a time frame corresponding to the industrial analogues, as learned from practical examples and geomechanical modelling results, i.e. critical anthropogenic-induced deformations (uplift or subsidence) which are accompanied with a drop of the minimal stress due to the acting extensional conditions. After this period, the stresses of the geological barriers may progressively change depending on the acting natural geological processes, which can be understood by the help of geological analogues for supporting argumentation with respect to the safety analysis of the long-term process.

In the past extensive analysis for several underground storage sites for chemo-toxic and radioactive wastes situated in salt formations have been performed for verification and validation as well as for the supporting arguments of the numerical geomechanical calculations, necessary for the integrity proof of the geological barrier. The experiences and results thus gained are available, representing natural analogues, sweeping over the industrial and geological time frame.

### **Discussion**

### **Isolation capability of salt rocks**

Salt formations are deemed to be impermeable. Whereon bases this special characteristic? On the one hand a salt rock is a polycrystalline chemical sediment that reacts with creep deformation due to its visco-plastic behaviour to slowly acting stresses without developing a joint system. On the other hand rock salt has a minor water content. Transport of mass only becomes possible from solid rock diffusion along grain boundaries without advective flow.

For nearly undisturbed or only weakly tectonically stressed salt rock structures it has been demonstrated that gases are fixed within the salt for at least 250 million years (Siemann, 2007). Investigations of Zechstein 2 (Stassfurt series) led to the result that methane within flatly bedded evaporites was stored at grain boundaries and inclusion nearly unaltered since sedimentation and diagenesis. Predominantly  $CH_4$  and  $H_2$  were detected at grain boundaries of rock salt from Zielitz (Germany). These investigation results represent a natural analogue for the fact that undisturbed salt formations are gas-tight geological barriers even for the very mobile hydrogen.

However, although gases are able to migrate from the adjoining rocks into the salt rocks due to tectonical processes such as volcanism or halokinesis, they can be fixed inside the salt (Schramm, 2007). An example for that is the salt deposit Werra/Fulda where inside the salt enormous amounts of  $CO_2$  are stored which penetrated into salt formations about 20 million years ago by magmatic intrusions during tertiary volcanism. The highest gas amounts occur in the so-called "Kristallbrockensalz" horizons – a very coarse-grained rock salt – within the middle Werra rock salt formation. The  $CO_2$  is trapped at inter-crystalline grain boundaries. During mining activities the stored  $CO_2$  can be released pneumatically.

Gas releases induced by blasting or drilling are frequent in the Werra potash district. The first major  $CO_2$  gas blow-out was triggered by an exploratory drilling occurred on 17 April 1958 and led to the  $CO_2$  pollution of the small mining field Menzengraben with six deaths. The biggest  $CO_2$  gas blow-out, caused by an exploration hole, occurred on 27-30 August 2003 in the potash mine Unterbreizbach. With a vertical hole drilled from a lower drift at about 950 m depth (situated in the Lower Werra rock salt) the thickness of carnallitite in the hanging wall was to be explored for a subsequent recovery of the carnallitite bulge. The vertical hole had been pierced through the 58 meters of carnallitite

and stood 4 meters in the Middle Werra rock salt when the blast was triggered. Through the exploration hole (37 mm diameter) enormous volumes of  $CO_2$  gas emptied into the pit which was estimated to be in the order of about 5 million cubic meters. Due to the cooling of the highly pressurised gas (near the lithostatic overburden pressure in the order of 20 MPa) at the exit of the hole, a 30 to 35 m long, 5-6 m wide and up to 3 m high  $CO_2$  glacier has been formed in the underlying rock salt drift, at an ambient temperature of about 30°C. This geological analogue (Figure 1) from the potash mining clearly demonstrates the isolation potential of rock salt for millions of years, even for trapped gases under high pressure.



Figure 1: CO<sub>2</sub> glacier after an underground gas breakout in salt rocks

On the basis of geological and, in addition, industrial analogues such as the practical experiences of high pressure gas storage in salt caverns over half a century, it can be concluded that undisturbed salt formations are impermeable!

## Loss of integrity and tightness of salinar barriers

Under undisturbed conditions, salt rocks are liquid- and gas-tight. Due to the visco-plastic properties of salt rocks, the undisturbed state in situ is characterised by nearly isotropic loading conditions. In accordance with Mohr's equations this means that the shear stresses at the grain boundaries of the polycrystalline salt rocks are nearly zero and the normal stresses are equivalent to the minimum principal stress  $S_{\text{MIN}}$ . Under these conditions, there is no interconnected pore space in the salt rocks. The porosity of salt rocks consists mainly of intercrystalline single cavities, i.e. isolated pores. A loss of tightness in the salt rocks can only be caused by the creation of connectivity, i.e. generation of interconnected pathways in the intercrystalline structures by:

- fluid-pressure-induced opening of grain boundaries and interconnection of intercrystalline flow paths (fluid-pressure-induced percolation);
- · deviatoric stress-induced crack growth and interconnection of intercrystalline cracks (dilatancy-induced percolation).

The two mechanisms for the generation of connectivity along the existing grain boundaries correspond to the generally applied criteria to investigate the tightness and integrity behaviour of salt rocks, which are also known as:

- · minimum stress or frac criterion (hydraulic criterion);
- · dilatancy criterion (mechanical criterion).

Salt rocks consist of crystal grains of varying geometry and texture. The discontinuities between the grains, i.e. the grain boundaries, represent a fluid-tight intergrown fracture network. The permeability of rock salt is due to the activation of this network by hydraulic pressure or deviatoric stress. The mechanically- or hydraulically-induced permeability of salt rocks results from the same micro-physical processes: the development of a flow paths network along grain boundaries after exceeding a threshold which is determined by the dilatancy boundary for deviatoric load conditions and by the acting normal stresses to compensate cohesive/adhesive grain bonds in the case of fluid pressure load.

Physically this means that percolation (Minkley, et al., 2012), i.e. the generation of interconnected flow paths, can be induced under certain conditions of normal stress and shear stress at the grain boundaries in polycrystalline salt rocks. Permeability in the salt rocks occurs when the tightness of the grain boundaries is lost, which can be caused by exceeding the two following threshold values:

- percolation threshold (1) under normal load with compensation of normal stress and contact cohesion/adhesion at grain boundaries;
- percolation threshold (2) under shear stress to overcome friction and cohesion/ adhesion at grain boundaries when the dilatancy boundary is reached.

Permeability is a sensitive indicator for the incipient opening of flow paths along grain boundaries of polycrystalline salt rocks, which are micro-mechanical weakness planes having a shear or tensile strength that is lower than the strength of the crystals. Based on this physical model concept an important statement for the tightness of saliferous barriers follows: a loss of tightness already occurs when one of the two mentioned percolation thresholds is exceeded, i.e. if either the dilatancy criterion or the minimum stress criterion is violated. Mechanical damage under deviatoric stress after exceeding the dilatancy boundary is basically relevant in the contour of an excavation, because its range into the surrounding rock is very limited. Most critical for the integrity of saliferous barrier is the minimum stress criterion, because it is not limited to the EDZ. In the following sections, this will be demonstrated by practical examples and industrial analogues from potash and rock salt mining.

## Loss of integrity of saliferous barriers due to convergence-induced static and dynamic loadings

Asse II (Germany)

An example of the violation of the dilatancy criterion due to convergence-induced deviatoric stresses, resulting in a loss of tightness of the geological barrier, is provided by the rock salt mining in steep inclined salt beddings at the southern flank of the Asse II. The load bearing system, comprised of slender pillars and only 6 m thick stopes, is not permanently stable dimensioned (Figure 2). Due to failures of stopes and the overload of slender pillars a significant convergence occurred which amounts to up to 6 m to date. The failure of the stopes has mainly contributed to the destabilisation of the mining system in the southern flank and to the large displacements (Figure 2). At the end of the 70 s this had already resulted in significant shear stresses along the margins of the mining areas. On the upper floor where the rock salt barrier thickness is lowest (less than 10 m), the dilatancy boundary has been exceeded.

Geomechanical calculations using a visco-elasto-plastic softening model (Minkley, et al., 2001 and Figure 10) facilitate a reproduction of the ongoing strain softening process in the load bearing system as characterised by a pronounced damage of rock salt barrier with a dilatancy in the order of > 10% at the 511 m level (Figure 3). In this area the micro-seismic activity is concentrated, indicating damage accumulation in the rock, which corresponds with the inflow zone.

Figure 2: Visco-elasto-plastic calculations for the damage processes in the load bearing system in the southern flank of the Asse

Left: Model section demonstrating the de-strengthening of stopes with development of shear zones over time. Top right: 3-D representation of the load bearing system with partially destroyed stopes. Bottom right: Fracturised stope in situ.

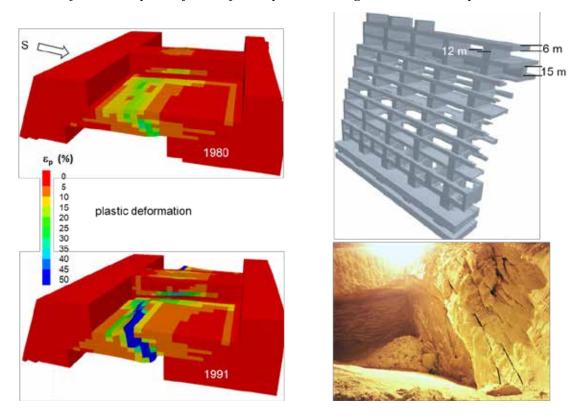
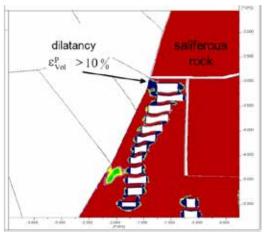


Figure 3: Calculated dilatant zones around the mining rooms at the southern flank of the Asse II mine (right) and recorded micro-seismic activities (left)

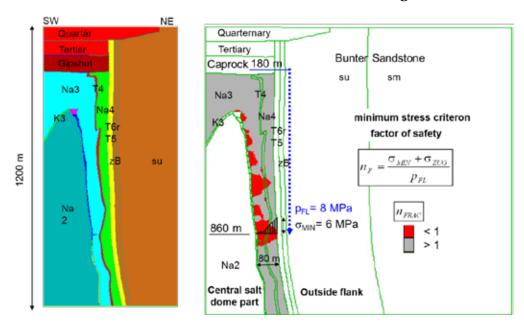




## Salt dome Bokeloh (Germany)

If strong mining-induced relaxation processes occur, the minimum stress criterion can be violated even under conditions of a thick saliferous barrier. In the salt dome Bokeloh (Figure 4) the sylvinite-bearing potash seam was mined by stoping, i.e. creation of steep inclined mining chambers exhibiting several meters thick slices of about 200 m height and 100 m width along the strike of the salt dome flank (Sessler & Holländer, 2002). Usually, the chambers should be backfilled with granular salt for stabilisation. Due to an inadvertently long open standing of mining chambers at 860 m depth a reduction of the minimum principal stress in the 80 m thick barrier (consisting of a sequence of Leine rock salt  $Na_3$ , red salt clay T4 and Aller rock salt  $Na_4$ ) was induced followed by water inflow from the salt dome edge. Thereby, it is important to note that the dilatant shear zones as computed with the visco-elasto-plastic material model, do not penetrate through the entire saliferous barrier, i.e. the dilatancy criterion is, at least, partly fulfilled. Thus, the later process cannot explain the water inflow.

Figure 4: Geomechanical calculations demonstrating the loss of tightness in the 80 m thick saliferous barrier at the salt dome edge Bokeloh



However, the supporting stress at the rim of the saliferous barrier, the Aller rock salt Na<sub>4</sub>, in a depth of 860 m amounts only to 6 MPa which is significantly lower than the measured fluid pressure values ( $p_{FL}$  = 8 MPa) in holes in the salt dome edge during pressure build-up test so that a fluid inflow became possible (Figure 4, right). As the main flow paths, delivering solutions from the salt surface (at the top of salt dome) down to 860 m depth, the steeply dipping discontinuities of the bedding are considered.

Currently, the transgressing fluids are collected through a hole, ranging from the mine to the outside edge of the salt dome, i.e. in front of the barrier, keeping the fluid pressure below the minimum principal stress, so that the inflow virtually came to a standstill. This demonstrates the potential of self-sealing in the salt if during pressure-driven percolation processes the liquid pressure falls below the minimum principal stress.

For this large-scale experiment (with convergence-induced lowering of the minimum principal stress at the edge of a salt dome under action of an existing fluid pressure), the important conclusion can be drawn that only an exclusive violation of the minimum stress criterion is sufficient to annihilate the barrier integrity.

### Weeks Island Mine (USA)

By mining in shallow depth with great subsidence and induced extension zones, there is the risk that the water pressure in the overlying rock will be greater than the minimum stress in the hanging salt barrier.

In the salt dome of Weeks Island rock salt has been mined at 150...220 m depth on two floors from 1902 to 1977 (Bauer, et al., 2000). The purpose of the rock salt mine was rededicated in 1981 as part of the Strategic Petroleum Reserve (SPR). After the storage of oil an inflow of water was noted, however, initially considered as non-threatening. In 1992, a sinkhole occurred and in 1995 a second sinkhole was found over the mine, and the oil has been drawn out again. Both sinkholes formed on mining edges, where the floors are stacked vertically and form a "hard mining edge" (Figure 5).

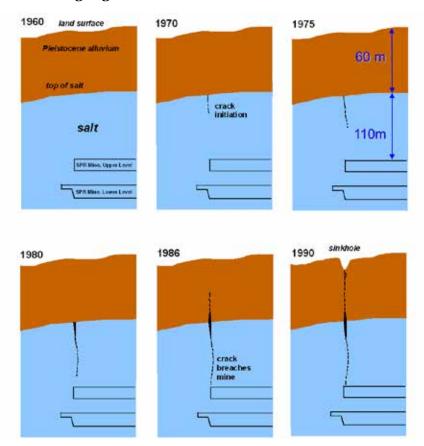


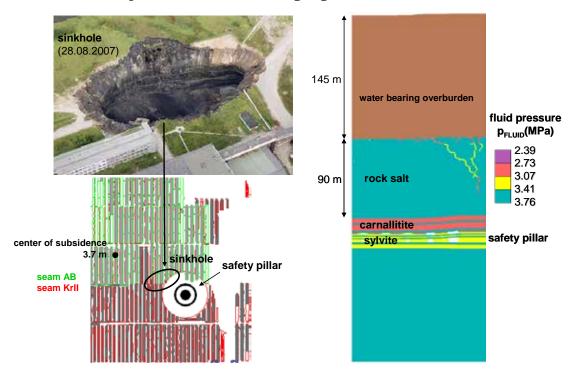
Figure 5: Sketch of the mechanism of fluid-pressure driven percolation above a mining edge of the Weeks Island Mine (after Bauer, et al., 2000)

Investigations which were performed to determine the causes with respect to geological peculiarities brought inconclusive results. Only after thorough investigation was the drowning of the mine linked to geomechanical causes. On mining edges extension zones in the overburden occurred in the 110 m thick overlying rock salt barrier due to the induced subsidence. This led to a decrease of the minimum principal stress in the area of the salt mirror where water pressures are acting. Once the minimum stresses in the rock salt dropped below the water pressure, grain boundaries in rock salt could be opened facilitating fluid-pressure-driven percolation of water pathways.

## Berezniki (Russia)

An analogous mechanism is responsible for the loss of integrity of the overlying geological saliferous barrier followed by drowning of sylvite mines Berezniki III (1986) and Berezniki I (2007) in flat bedding. Similar to that what happened in the Weeks Island Mine, a sinkhole occurred above the Berezniki I mine close to a hard mining edge, which developed where the mining areas of two floor terminated at the downhole safety pillar. Northwest at the same location, the largest subsidence was measured above the Berezniki I salt mine in the order of 3.7 m (Figure 6).

Figure 6: Sinkhole Berezniki I and coupled hydro-mechanical calculation with fluid pressure-driven generation of hydraulic flow paths above a hard mining edge in the extension zone



The IfG conducted a back analysis for explaining the integrity loss on the basis of coupled hydro-mechanical calculations. The geomechanical model describes the situation of the two floor mining in about 250 m depth in the sylvinitic seam Kr II and the partially carnallitic seam AB using the visco-elastic-plastic material model for saliferous rocks (Minkley & Mühlbauer, 2007).

The geomechanical model demonstrates the development of an extension zone above the hard mining edge as a result of the convergence and subsidence incidents, so that the minimum principal stress drops below the water pressure in the overburden and pressure-driven hydraulic flow paths are generated in the 90 m thick overlying saliferous barrier (Figure 6). After the time dependent breakthrough along the pressure-driven opened grain boundaries flow paths were developed into the dilatant zone surrounding the mining openings which are followed by dissolution processes in the saliferous strata. Finally, the resulting cavity in the salt became so large that the overlying overburden could no longer resist and a sinkhole was formed (Figure 6).

## Völkershausen (Germany)

In the potash mine Merkers in Thuringia a globally unique natural analogue exists for the gas frac of a geological rock salt barrier with subsequent healing. During the dynamic high loading caused by the rock burst Völkershausen (1989) (magnitude ML=5.6), the lying rock salt barrier had lost its integrity due to special conditions as shown below. In the hanging wall the greater thickness and the layered structure forming a geological multi-barrier system (consisting of approximately 180 m rock salt and 40 m salt clay and claystone) was advantageous, i.e. due to its protective capacity no overall integrity loss of the hanging barrier occurred (Figure 7).

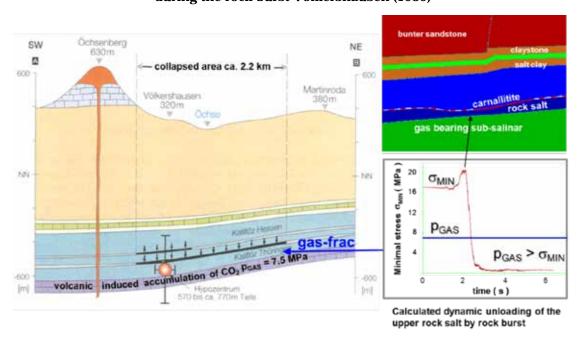


Figure 7: Gas-frac in the underlying salt barrier during the rock burst Völkershausen (1989)

In the area of the Feldatal fault, associated with the laminar thinning of the Lower Werra rock salt to less than 30 m below the eastern edge of the rock burst field, the minimum stress criterion was violated. Due to the dynamic, convergence jump occurred during the collapse of the mining field. With respect to the failure of the underlying barrier the dynamically induced stress redistribution below the eastern edge of the collapse field was of utmost importance, where the chain-reaction-like pillar fracture process came to a halt. After the pressure wave in front of the fracture front associated with the convergence step in the mining horizon passed through, the principal minimum stress at the base of the Lower Werra rock salt dropped abruptly from about 18 MPa to a few MPa (Figure 7) in the thinned area below the eastern edge of the rock burst field, as demonstrated by the dynamic calculation (Minkley, 2004). The subsaliniferous bed had been used for decades for the extraction of volcanic CO<sub>2</sub>, which accumulated during basaltic volcanism 15 to 20 million years ago in the Vorderrhön region of Germany. In drillings, a CO<sub>2</sub> gas pressure in the order of 7.5 MPa was observed.

From the calculated stress field after the rock burst it becomes clear that the safety factor against a tearing of the underlying rock salt barrier lies far below 1 in the area of thinning of the Lower Werra rock salt. Due to the acting gas pressure at the base of the Lower Werra rock salt in the specified magnitude, it inevitably came to a gas fracture due to the dynamically occurring strong relaxation in that area.

Immediately after the rock burst at the underground sites 19 and 20 at the southeast rock burst field a gas outflow of  $CO_2$  was observed. Until the end of 2000 the total gas outflow into the underground openings was around 46 million  $m^3$   $CO_2$ .

As part of a research project (Popp, et al., 2007) the condition of the underlying rock salt barrier, which became leaky by the gas frac due to the rock burst in 1989, has been investigated. In the 240 m long horizontal core drilling well Hz 156 situated in the Lower Werra rock salt in the region of the former  $CO_2$  breakthrough zone we found healed and partly open gas fractures in the area (Figure 8). The orientation of the cracks corresponds to the calculated direction of the major principal stress. Eighteen (18) years after the rock burst, hydro frac measurements showed a stress build-up to  $s_{\rm MIN} > 11$  MPa in the field of the former gas frac zone in the Lower Werra rock salt, which is higher than the acting  $CO_2$  gas pressure.

On the basis of investigation and geomechanical analysis results, the decision was taken to finish the extraction of  $CO_2$  and brines from the Lower Werra Anhydrite (A1) which was started after the rock burst. The production wells Hz 154/99 Hz and 143/4 (Figure 8) were closed on 16 February 2007. It has to be mentioned that the lying rock salt barrier has regained its tightness due to the specific properties of rock salt with the reactivation of cohesive/adhesive forces on crack interfaces while reducing the surface energy (Minkley, 1989), i.e. self-sealing and healing of cracks.

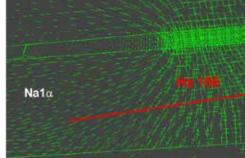
This unique large in situ experiment demonstrates the recovery of the barrier integrity within a rock salt formation after the occurrence of a gas frac induced by dynamic unloading, which undoubtedly confirms the potential for self-sealing of rock salt.

This example is important for the assessment of the so-called gas frac scenarios for a repository in rock salt. In the post-closure phase of a repository, the presence of moisture results in the corrosion of the metal waste containers, which will lead to the formation of hydrogen. Due to the gas tightness of the rock salt, there is concern that due to a pressure increase the barrier integrity could be compromised by a gas frac (RSK, 2005).

Figure 8: Drilled gas frac zone in the Lower Werra rock salt and orientation of gas frac cracks in the direction of the major principal stress

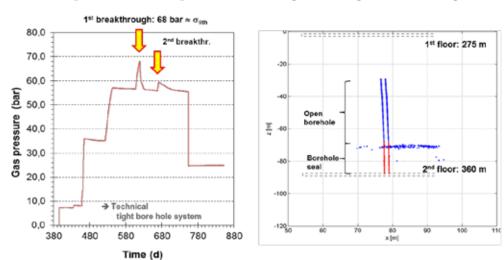


- At least partial, but "real" healing is documented
- Only local gas reservoirs were found without connection to the gas bearing sub-salinar
- Recovery of minimal stress in the former gas-frac zone
- Gasfracs are oriented parallel to the direction of the maximal stress



To investigate this question a large scale pressure test (with a 50 m³ gas volume) was designed to proof already existing results from small scale bore hole tests (only 0.05 m³) (Popp, et al., 2007). In the German salt mine Merkers a 60 m long vertical wellbore (blind hole – with a diameter of 1.30 m) was drilled in the Middle Werra rock salt zone in a depth of 300-360 m. The test site was surrounded by a highly-sensitive micro-seismic network (operated by GMUG) allowing the on-site monitoring and localisation of local micro-crack events (local resolution in the order of some decimetres). After sealing the lower bore hole end with a 20 m long gas-tight plug (constituted of a special MgO-concrete) the gas volume was pneumatically pressurised in numerous steps (Popp, et al., 2012).

Figure 9: Course of the gas pressure build-up in the wellbore (left) and vertical projection of the localised acoustic events (right) with pressure-driven percolation during the first gas breakthrough



Reaching a maximum pressure of 68 bar (which is in the order of the minimum stress), an increase of micro-seismic activity was observed in a very narrow spatial horizon oriented parallel to the bedding. The accumulated local micro-seismic events correspond to a 15 m wide eastward ranging lateral AE-zone of about 2 m thickness which gradually developed over a period of 3 days after the pressure increase (Figure 9). Then a fluid outflow on discrete flow paths into two of the open holes of the AE arrays was recognised. The fluid-breakthrough is associated with a pressure decay stabilising at around 57 bar. This clearly documents the self-sealing capacity of salt after a gas breakthrough, because of the reactivation of cohesion (Minkley & Groß, 1988) as it is shown by the healed gas-fractures after the rock burst Völkershausen (Figure 8). It is also advantageous, that the gas breakthrough process is repeatable.

With respect to the acting physical mechanism, it has to be taken into account that, in contrast to liquids, gases are compressible and have a much higher viscosity, so that upon reaching the minimum principal stress in the salt formation, a dynamically-running pressure-driven percolation process (local fracturing and opening of pathways on grain boundaries) is initiated, since in spite of crack opening, unlike liquids, no pressure drop at the crack tip occurs due to the underlying larger volume of gas. In accordance with the percolation threshold (1) this corresponds to micro-fracturing of cohesive bonds on grain boundaries at the crack tip by compensation and overcoming of the normal stress.

Unfortunately, no drilling was performed into the potential gas breakthrough zone, i.e. no core information about the real nature of this zone is available. Thus, the range and mechanism of gas percolation must be implicitly checked by further studies. It can

be assumed that the range of gas percolation with respect to the length of gas fracs depends on the existing gas volume.

A geological analogue for localised gas fracs in salt exists as given by the so-called "slices", i.e. very sharp fractures of less than 1 cm width, which are mostly healed but still visible ranging over several decametres. They are created in the course of tertiary volcanism in the Werra potash area, partly still carrying  $CO_2$  gas, which gives hints that these gas fracs occurred when the salt was dynamically pressurised.

In the case of gas or liquid pressure on salt formations under static or dynamic load conditions a directional percolation develops as soon as the percolation threshold (1) is exceeded. The fluid pressure-driven percolation occurs along discrete flow paths generally in the direction of the maximum principal stress under inclusion of bedding plans and the lithological microstructure.

## Barrier integrity under thermo-hydro-mechanical loading

With respect to the disposal of heat-generating radioactive waste in a repository mine the geological barriers surrounding the underground openings are firstly affected by convergence, as it is the same case in a conventional salt mine. After emplacement of HAW materials a temperature increase occurs due to the thermal impact by the radioactive decay resulting progressively in an uplift process as a consequence of the rock expansion in the hanging layers. Regarding the consequences of displacements which may be accompanied with a decrease of the restraining stresses below the acting fluid pressures there is no difference from the mechanical point of view whether they are caused by subsidence or elevation processes. However, in the case of heat-generating waste the barrier integrity has to be investigated under the action of internal and external hydro-thermo-mechanical stresses.

The pressure-driven generation of flow paths in a salt dome may preferably act on bedding surfaces and discontinuities in the various saliferous units (Figure 9). Thus, steeply dipping bedding planes and discontinuities in salt domes which extend in upper groundwater bearing areas of the salt surface level, if no overlaying clay layers exist, are always potential weakness points in the geological barrier, because along discontinuities, a preferred fluid transport is possible (Grundfeld, et al., 2005).

Boundaries at interfaces between neighbouring salt layers (lithological boundaries) and discontinuities within salt portions (e.g. due to bedding planes), which result in different mechanical properties including thermal expansion coefficients, elastic and plastic properties as well as creep properties, are potential predominant pathways as a result of stress differences and slipping processes upon thermo-mechanical stresses. The pressure-generated networking of hydraulic flow paths along discontinuities on micro- and macroscopic scale is of decisive importance for the tightness of salinar barriers at thermo-mechanical loading, if the minimum principal stress drops below the fluid pressure at the salt top due to the thermal expansion and uplift. Mechanical damage after exceeding the dilatancy boundary is limited to the immediate vicinity of the excavations, representing the EDZ, which is only of secondary importance for the overall integrity and tightness of the salinar barrier. Due to the thermal load of the radioactive waste and the following thermal expansion, additional stresses are induced in the surroundings of the storage horizon. These lead to uplifts in the overlying layers up to the surface, which are in the order of meters.

To demonstrate the integrity of the geological barrier under the thermo-mechanical stresses the complex mechanical behaviour of the general suite of saliferous rocks has to be described, i.e. ranging from ductile and creep (like rock salt) via brittle and creep (typical of carnallite) to brittle without creep (as anhydrite) (Figure 10). In addition, the mechanical and hydraulic behaviour of the lithological interfaces and discontinuities has to be taken into account by a discontinuum mechanical approach (Minkley & Mühlbauer, 2007).

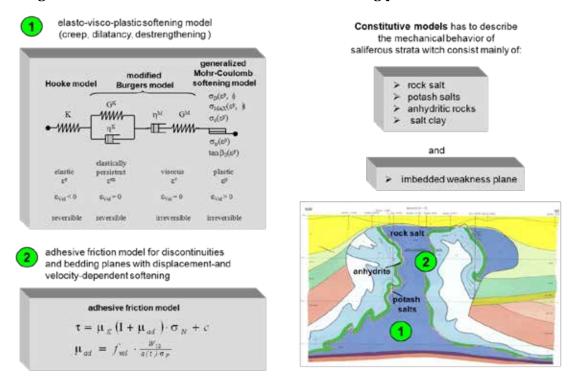


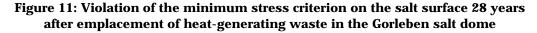
Figure 10: Material models for saliferous rocks, bedding planes and discontinuities

In the framework of the preliminary safety analysis of Gorleben, comprehensive thermo-mechanical calculations were carried out. The calculation results convincingly show the general mechanisms that can lead to a temporary local violation of the tightness and integrity of the geological barrier: Due to the heat supply and the accompanied uplift the state of stress is spaciously altered in the salt dome. At the upper salt regions a reduction of the minimum principal stress occurs (thermo-mechanical stress drop) due to the volume expansion of extensive rock portions. The relaxation area, where the stress drops below the acting brine pressure and where a fluid pressure-driven percolation can occur due to the violation of the minimum stress criterion, ranges a few hundred meters below the salt level in the model calculations (Figure 11). A fluid pressure-driven percolation is also documented in the thermo-mechanical calculations along the steeply dipping bedding planes between "Streifensalz" z2HS2 and "Kristallbrockensalz" z2HS3 and along the existing lithological boundary between "Kristallbrockensalz" and carnallitite (potash seam, z2SF).

As the main outcome of the geomechanical analysis it has been concluded that, based on the evaluation of the minimum stress criterion, as done in the thermo-mechanical calculations, under quasi-static loading conditions no persistent violation of the integrity of the salt portions between the salt levels and the emplacement horizon is observed.

The strongest thermo-mechanical impacts of heat-generating waste storage on the barrier integrity occur some decades after the emplacement process (Figure 11). In the subsequent time periods, the areas in which a violation of the minimum stress criterion was observed will continuously decrease as a consequence of the always acting creep processes, resulting in a stress re-distribution, even if the salt dome uplift proceeds due to the heat supply.

In Figure 12, the thermo-mechanically calculated time evolution of the minimum principal stress after storage of heat-generating radioactive waste is shown for two points in the overlying saliferous barrier. The drop of the minimum principal stress, as induced by volume expansion, inducing extensional conditions due to the uplift of the domal structure,



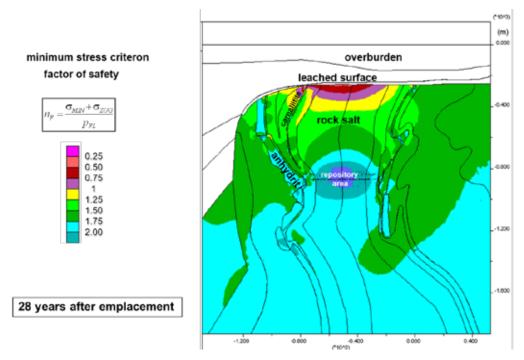
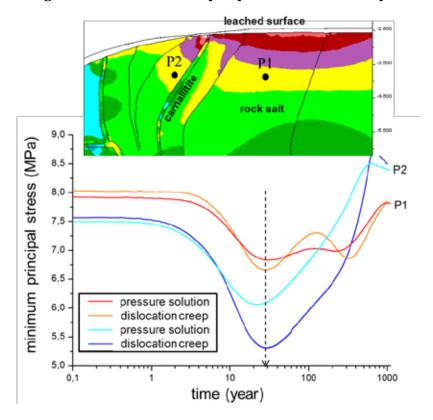


Figure 12: Time course of the decrease of the minimum principal stress for two points in the saliferous barrier after emplacement of heat-generating radioactive waste, taking either a dislocation creep or pressure solution creep into account



reaches its minimum 30 years after emplacement. The visco-elasto-plastic calculations have been performed for two different creep law approaches. The first adaptation is based on the stationary approach BGRa, a power law which describes the dislocation creep in the salt crystals. The second approach takes into account the grain boundary effects, which may happen if at lower deviatoric stresses fluid films are present at the grain boundaries, i.e. the pressure solution creep may lead to higher creep rates (Urai & Spiers, 2007). With the visco-elasto-plastic constitutive model of IfG both creep mechanisms can be described with the sine-hyperbolic creep approach as implemented (Figure 13). The calculations show that if the pressure solution creep is considered due to the resulting higher creep rates at lower deviatoric stresses the decrease of the minimum principal stress and thus the temporary integrity violation of the saliferous barrier is lower (Figure 12).

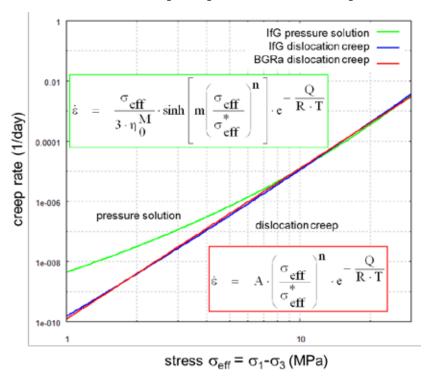


Figure 13: Stationary creep laws for rock salt for dislocation creep and pressure solution creep

As an outcome of the thermo-mechanical calculations it has to be noted that the largest hydraulic stresses on the geological barrier have to be expected in the first hundred years of the repository use (Figure 12). That is to say, in the first hundred years, the stress of the geological barriers by anthropogenic intervention is greatest. After this period, a loss of integrity of the saliferous barrier due to pressure-driven inflow of groundwater (by percolation) is no longer possible. Again the natural geological barrier behaviour will become progressively dominated by that influence factors which have preserved the integrity of the entire salt deposits over hundred millions of years.

For the Gorleben site the integrity of the saliferous barriers under dynamic stresses (earth quakes) was investigated with the macro seismic location intensity of 7.3 (MSK), corresponding to a magnitude M=4.5, as derived from the design earthquake (NLfB, 1996). The earthquake excitation was simulated at the time when the drop in the minimum principal stress reaches deepest in the saliferous barrier, i.e. after about 30 years, as predicted by the static modelling results reported above. The subject of the simulation of the earthquake impact was, in particular, the hydraulic behaviour of discontinuities and bedding interfaces, which were affected by brine pressures acting in the areas of the salt

surface. In coupled hydro-mechanical calculations it was demonstrated that during the passage of seismic waves with a maximum acceleration of  $1.4~\text{m/s}^2$ , the salt water occurring at the salt surface may penetrate pressure-driven about 30 meters deeper into the steeply dipping discontinuities than under static conditions (Figure 14).

overburden

| Same before earthquake | Substitution | Substitution

Figure 14: Pressure-driven percolation of brine present at the salt surface in a steeply dipping discontinuity during dynamic conditions (earth quake) of the Gorleben salt dome

#### **Conclusions**

Geological analogues undoubtedly demonstrate that salt formations in the undisturbed state are liquid and gas tight under isotropic stress conditions over geological times. It has to be shown in geomechanical calculations that the same holds generally for the integrity of the salt barrier in the case of the final disposal of heat-generating radioactive waste, where the minimum principal stress may be locally lowered (thermo-mechanical stress drop).

Of special importance, as shown by the performed numerical calculations, is that already some few decades after the emplacement of heat-generating radioactive waste the largest drop in the minimum principal stress occurs in the overlying salt barrier. After this relatively short time period of high loading the stress state develops back in the direction of the isotropic initial stress state due to creep processes in the visco-plastic reacting salt rock, although the uplift process will proceed for a long time.

The critical condition of the barriers loaded by volume expansion in the host rocks surrounding the repository lies in a time window for which numerous examples from the potash and rock salt mining industry exist where a loss of barrier was caused by subsidence and associated relaxation processes. Thus, these industrial analogues are of great practical importance for the assessment of barrier integrity of geologic waste repositories.

In conventional potash and rock salt mining several failure cases are known where a loss of integrity and tightness of the geological barriers in the first hundred years of excavation occurred. These industrial analogues, as discussed before, have to be considered in the general assessment of barrier integrity of geologic waste repositories.

The back analysis of failure and flooding events in potash and rock salt mining, as to be done in coupled mechanical-hydraulic models, is advantageous to validate the models itself and is, therefore, essential for a reliable forecast for assessment of barrier integrity, if salt repositories are used for the disposal of radioactive waste. The models are qualified to describe future developments of integrity behaviour reliably only if the failure mechanisms can be plausibly demonstrated by geomechanical models using suitable material approaches.

As learned from drowning events the hydraulic pressure acting at the cap rock usually exceeds the minimum principal stress in the rock salt. This is the case when above extraction edges around mining fields the restraint is time-dependently reduced by the developing subsidence processes and extensional loadings, the minimum stress becoming lower than the acting ground water pressure.

The formation tightness, which is a distinguished characteristic of salt rocks, is lost when one of the percolation thresholds, the minimum stress or the dilatancy criterion is exceeded. While dilatancy takes place in the immediate cavity surroundings, i.e. the EDZ, where the deviatoric stress is high, the stress changes with a violation of the minimum stress or hydraulic frac criterion may affect more extended barrier areas. The physical background for the action mechanism of the minimum stress criterion is primarily the discontinuum-mechanical structure of rock salt on micro-mechanical scale as a polycrystalline solid with lower strength at the grain boundaries. The acting mechanism, i.e. a loss of the tightness through fluid pressure-driven percolation is well proven by experimental tests in the laboratory and at large scale. Connectivity along the grain boundaries of the salt crystals is established when the normal stress acting on the grain boundaries is compensated by the fluid pressure.

With respect to the disposal of chemo-toxic and radioactive waste in salt formations, especially the reliability of sealing properties of salt rocks is of utmost importance if the principle of complete enclosure of the waste from the biosphere is required. The proof of the tightness and integrity has to be performed under consideration of the geogenic and anthropogenic loadings affecting the geological salt barrier.

With respect to coupled hydro-mechanical processes macro-discontinuities such as bedding planes or lithological interfaces also have to be considered in addition to the discontinuum-mechanical pore space structure of the salt rock on the micromechanical level. This is always the case if complex structures like salt domes should be investigated regarding their response under the action of inner thermo-mechanical and external hydraulical loadings during the disposal of radioactive waste. For the tightness of the saliferous barriers under thermo-mechanical stresses the fluid pressure driven percolation of hydraulic flow paths along discontinuities in the micro- and macroscopic scale (grain boundaries, bedding planes) is of utmost importance, especially in the case of a drop of acting stresses below the fluid pressure present at the salt surface due to the thermal rock expansion and salt dome uplift.

A violation of the minimum stress criterion can result again from extensional stress regimes, however, in this case due to uplift processes, in contrast to drowning events of a mine which are induced by subsidences. Nevertheless, from a mechanical point of view there is no difference between these reasons. Crucial for the integrity of the hydraulic saliferous barrier is only whether the minimum stress criterion is violated or not. The higher the creep capability of the solid salt rocks, the lower a possible loss of integrity in the saliferous barrier.

For the establishment of sufficient barrier thickness for a geological repository the experiences from industrial analogues should be used (Minkley, 2009). For the barrier integrity under long-term conditions, i.e. where nearly isotropic stress conditions are acting in the salt the geogenic processes are crucial, which are documented and supported by geological analogues.

Finally, based on thermo-hydromechanical calculations an undisturbed, sufficiently thick salt barrier – the so-called isolating or containing rock zone – around a geological HAW repository has to be demonstrated, where neither the minimum stress nor the dilatancy criterion is violated. This has been done analogous for the geomechanical integrity proof for chemo-toxic disposal sites, potash and rock salt mines or gas storage caverns situated in salt formations.

With respect to the principle of complete containment, salt formations have unique advantages for a nuclear waste repository:

- · impermeability in undisturbed conditions;
- tightness will only get lost in case of exceeding a threshold;
- · self-sealing ability and healing by reactivation of cohesive/adhesive bonds on fractures because of its visco-plastic behaviour.

Finally, it can be stated that Germany offers excellent geological conditions in domal as well as bedded salt formations for final disposal of HAW waste. However, a simple structure and robust geological multi-barrier system that combines the advantages of salt and clay formation (natural multi-barrier system) is most likely to be found in flat bedded salt at comparatively lower exploration costs.

## References

- Bauer, S.J., B.L. Ehgartner and J.T. Neal (2000), "Geotechnical Studies Associated with Decommissioning the Strategic Petroleum Reserve Facility at Weeks Island, Louisiana: A Case History", SMRI Fall 2000 Meeting, San Antonio, Texas, USA, 15-18 October 2000.
- Bergmannsverein (2002), 150 Jahre Salzbergbau Stassfurt Wiege des Kalibergbaus, Salzland Druck Staßfurt GmbH & Co. KG.
- Eickemeier, R., et al. (2012), "Preliminary Safety Analysis of the Gorleben Site: Thermomechanical Analysis of the Integrity of the Geological Barrier in the Gorleben Salt Formation", WM2013 Conference, Phoenix, Arizona, USA, 24-28 February 2013.
- Grundfelt, B., et al. (2005), "Sicherheitstechnische Einzelfragen Bedeutung des Mehrbarrierenkonzepts für ein Endlager für radioaktive Abfälle beim Nachweis der Einhaltung von Schutzzielen", Kemakta Konsult AB-AR 2005-28.
- Minkley, W. (1989), "Festigkeitsverhalten von Sedimentgesteinen im post-failure-Bereich und Gebirgsschlagerscheinungen", Proc. Int. Symp. Rock at Great Depth, Pau, Vol. 1, pp. 59-65.
- Minkley, W. (2004), "Back Analysis Rock Burst Völkershausen 1989", Proc. 1st International UDEC/3DEC Symposium, Bochum, Germany, 29 September-01 October 2004, Tayler & Francis Group, London, ISBN 90 5809 6361, 105-112.
- Minkley, W. (2009), "Final Disposal in Rock Salt and Barrier Integrity", Proceedings of the European Commission TIMODAZ-THERESA International Conference, Luxembourg, 29 September-01 October 2009, European Union 2012, ISBN 978-92-7926057-5, 105-121.

- Minkley, W., M. Knauth and U. Wüste (2012), "Integrity of Salinar Barriers Under Consideration of Discontinuum-Mechanical Aspects", Mechanical Behavior of Salt VII, Paris, France, 16-19 April 2012, Taylor & Francis Group, London, ISBN 978-0-415-62122-9, pp. 469-478.
- Minkley, W. and J. Mühlbauer (2007), "Constitutive Models to Describe the Mechanical Behavior of Salt Rocks and the Imbedded Weakness Planes", in M. Wallner, et al. (Eds.), The Mechanical Behavior of Salt Understanding of THMC Processes in Salt, 6<sup>th</sup> Conference (SaltMech6), Hannover, Germany, 22-25 May 2007, Taylor & Francis, pp. 119-27.
- Minkley, W., et al. (2001), "A Visco-Elasto-Plastic Model and Its Application for Solving Static and Dynamic Stability Problems in Potash Mining", Proc. 2<sup>nd</sup> Int. FLAC Symposium, Lyon, France, 29-31 October, pp. 21-27.
- Minkley, W. and U. Groß (1988), "Zum Haftreibungsverhalten an Gesteinstrennflächen in Abhängigkeit von der Belastungsgeschichte", Felsbau, 6, Nr. 4, 184-188.
- NLfB (1996), Stellungnahme zu dem "Seismologische Gutachten" erstellt von der Abteilung Erdbebengeologie des Geologischen Instituts der Universität zu Köln, Niedersächsiches Landesamt für Bodenforschung, Hannover.
- Noseck, U., W. Brewitz and I. Müller-Lyda, (2008), "To What Extent Can Natural Analogues Contribute to the Safety Case of High Level Waste Repositories in Rock Salt?" Safety Cases for Deep Geological Disposal of Radioactive Waste: Where Do We Stand? Symposium Proceedings, Paris, France, 23-25 January 2007, NEA-No. 6319, ISBN 978-92-64-99050-0.
- Popp, T., et al. (2007), Untersuchungen zur Barriereintegrität im Hinblick auf das Ein-Endlager-Konzept, Institut für Gebirgsmechanik GmbH, Leipzig, Forschungsvorhaben SR 2470.
- Popp, T., et al. (2012), Auswirkungen der Gasbildung im Endlager auf den einschlusswirksamen Gebirgsbereich, Forschungsvorhaben (FKZ) 3609R03222, Institut für Gebirgsmechanik GmbH, Leipzig.
- Reaktor-Sicherheitskommission (RSK) (2005), Stellungnahme Gase im Endlager vom 27.01.2005 (379. Sitzung).
- Schramm, M. (2007), "Vorkommen natürlicher Gase im Salz", Workshop der GRS, Berlin, 17-18 April 2007, GRS-242, pp. 157-166.
- Sessler, W. and R. Holländer (2002), "Das Kaliwerk Sigmundshall der K+S AG", Akad. Geowiss, Hannover, Veröffentl, 20, 70-76.
- Siemann, M.G. (2007), Herkunft und Migration mineralgebundener Gase der Zechstein 2 Schichten in Zielitz, Kali und Steinsalz, Heft 3/2007, 26-41.
- Urai, J.L. and C.J. Spiers, (2007), "The Effect of Grain Boundary Water on Deformation Mechanisms and Rheology of Rock Salt", in M. Wallner, et al. (Eds.), The Mechanical Behavior of Salt Understanding of THMC Processes in Salt, 6<sup>th</sup> Conference (SaltMech6), Hannover, Germany, 22-25 May 2007, Taylor & Francis, pp. 149-158.

## NEA/RWM/R(2013)10

## Internal dynamics of salt structures<sup>1</sup>

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In the literature salt structures are represented in two strikingly different ways. In studies from hydrocarbon exploration and producing using 3-D seismic and well data that focus on the sub- or suprasalt sediments, the evaporites are shown as homogeneous bodies. On the other hand, studies of the internal structure of salt show the extremely complex internal geometry with much less attention to the structure of the surrounding sediments. Most current numerical and analogue models of salt tectonics also tend to assume relatively simple internal structures. New developments in microstructure analysis, combined with 3-D seismic study and modelling of complex internal structures in salt form the basis of integrating these two.

A review and synthesis of the mechanical and transport properties and their extrapolation to relevant strain rates must be based on an understanding of the microscale deformation mechanisms in natural laboratories and measurement of salt flow in situ. Dislocation creep and grain boundary dissolution-precipitation processes, such as solution-precipitation creep and dynamic recrystallisation, both play a significant role and grain boundary healing in deforming salt may cause major changes in rheology, at time scales both relevant to geologic evolution and subsurface operations. New methods of microstructure analysis based on microstructure decoration, orientation analysis and trace element geochemistry, combined with paleorheology indicators based on structures observed in natural laboratories allows an integration of these data and the development of a unified model for salt creep for both underground cavities and natural deformation. The model also includes the effect of high fluid pressures in salt which lead to dramatic increases in permeability, strongly reducing sealing capacity.

Many evaporate deposits contain brittle-ductile claystone and/or anhydrite layers enclosed in salt. Although these stringers can be reservoirs for hydrocarbons and can pose serious operational challenges, little is known about the early evolution and deformation history of these layers. Three-dimensional (3-D) seismic study of these, combined with well data and core analysis of diagenetic evolution shows highly complex structures caused by both brittle and ductile deformation, in good agreement with observations in salt mines and analogue models. These form the basis of a new generation of mechanical models to predict the complex coupling between the internal deformation of the salt and the evolution of the surrounding sediments.

197

<sup>1.</sup> The full paper being unavailable at the time of publication, a slightly adapted version of the contribution as presented at the symposium is provided here.

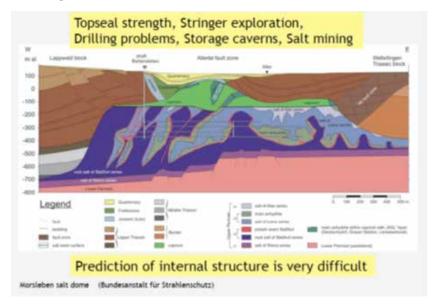
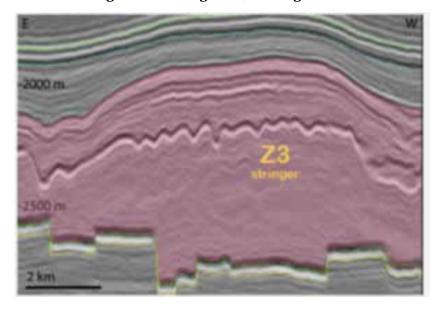


Figure 1: Internal structure of salt bodies - who cares?

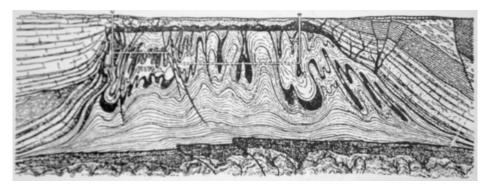




Source: H. van Gent.

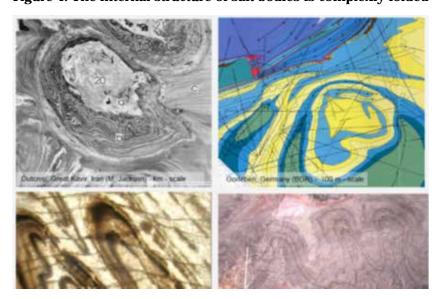
Figure 3: Internal structure

Layered evaporites with K-Mg salts and anhydrite



Source: Abb, 164, Schematisches Profilbild eines Salzstocks (nach E. Seidl).

Figure 4: The internal structure of salt bodies is complexly folded



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Figure 5: Z3 stringers

Source: H. van Gent, 2009.

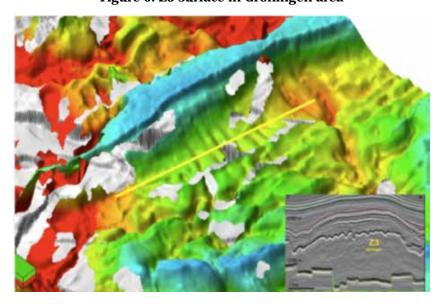
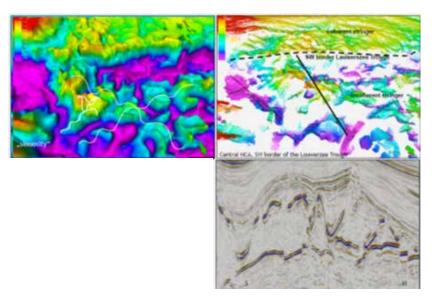


Figure 6: Z3 surface in Groningen area

Source: H. van Gent, 2009.

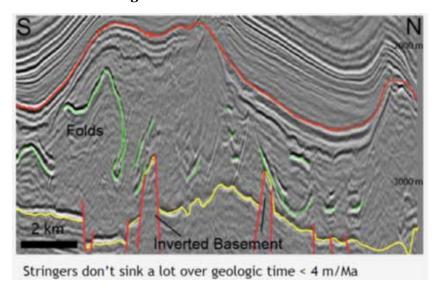
Figure 7: Incoherent stringer

F2 + F3 folds



Source: F. Strozyk.

Figure 8: Western offshore area



Source: H. van Gent, 2009.

Stephase 2nd phase 3rd phase

Early - Middle Lints Triansis - Present day 2007 0

Name of the Stephase Stephase

**Figure 9: Prediction of internal structure - Step 1** 

Source: Mohr, et al., 2004.

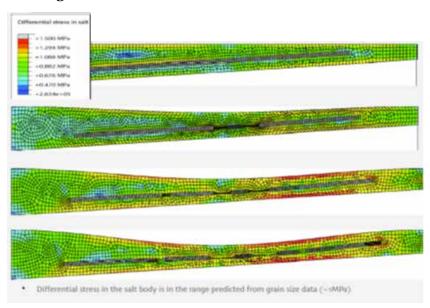


Figure 10: Model evolution - differential stress in salt

Source: Shiyuan Li.

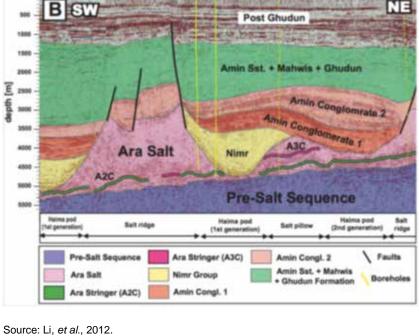
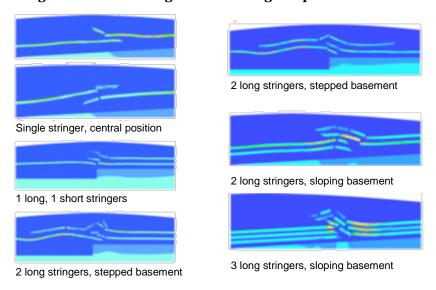


Figure 11: Stringers in Ara salt

Figure 12: Brittle stringer in deforming salt pillow - overview



Source: Shiyuan Li.

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Figure 13: Viscous stringer in deforming salt pillow

Source: Shiyuan Li.

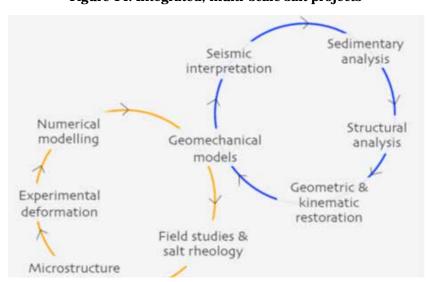


Figure 14: Integrated, multi-scale salt projects

## **Session IV**

**Long-Term Properties of Geotechnical Barriers** 

## NEA/RWM/R(2013)10

# Geotechnical barriers in the repository concept for HLW disposal in rock salt

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Plugs and sealings play an important role in the safety concept for repositories in rock salt. The sealing design for the German safety concept is based on a refilling of the open void volumes in the emplacement areas after disposal of the waste with crushed salt. The cross drifts of the disposal areas will be sealed with dams against the main drifts and the transport drifts with sealings against the infrastructure area. Shaft sealings are needed to prevent water inflow through the shafts. Some analogues addressing selected aspects for the long-term behaviour of such geotechnical barriers have already been identified. However, an information exchange with industry might help to find additional "industrial" analogues to be used in repositories in rock salt.

#### Introduction

Besides the geological barrier the major safety function "containment of the waste" will be provided during the first phase after closure by the shaft and drift sealings. In a later phase compaction of the backfill will lead to a very low permeability and take over this safety function. The layout of the geotechnical barriers will be done in a way that by coaction with the geological barrier an inflow of solutions to the waste will be avoided or at least highly reduced, preventing a significant release of contaminants out of the repository (Mönig, et al., 2011).

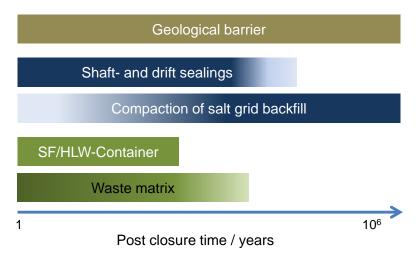
Therefore, arguments from analogue studies can be important to support statements that the hydraulic resistance of geotechnical barriers is high enough for the required time frame to avoid water flow into the repository. Additionally, analogues demonstrating a compaction of crushed salt with time leading to low porosities and permeabilities are of high relevance for the safety case.

The assessment of the integrity of the geological and geotechnical barriers is a key aspect of the safety and assessment concept for a high-level waste repository in Germany. Three aspects are of high importance regarding the geotechnical barriers:

- 1. hydraulic resistance of the barriers;
- 2. long-term stability of materials;
- 3. compaction behaviour of crushed salt.

The largest difficulties result from the fact that the laws for the sealing of excavation disturbed zones or for the compaction of crushed salt are not fully assured. This is because complex processes in the applied materials are partly described by simplified models and some of the parameter values not always are precisely enough known. The evolution of the geochemical conditions is also difficult to predict, which causes uncertainties with regard to the long-term stability of sealing materials.

Figure 1: Barriers of the repository system in rock salt potentially contributing to the safety function "containment of the waste" and time frame they are active according to Mönig, et al. (2012) (not to scale)



#### **Discussion**

## Hydraulic resistance of geotechnical barriers

The hydraulic resistance of sealings is an integral resistance of excavation disturbed zone (EDZ), seal body and the contact zone between the underground structure and the rock salt. One interesting analogue with respect to this topic is the permeability reduction of the excavation disturbed zone in rock salt observed in an old drift in the Asse mine. The primary reason for EDZ formation is the stress imposed by excavation. It leads to dilatancy which results in an increase in porosity and permeability in a limited zone around the drift wall and the drift floor. Natural undisturbed rock salt is impermeable for fluids because of its low permeability of approximately  $10^{-21}$  m². The permeability of the excavation disturbed zone can be increased to values of  $10^{-15}$  m². Therefore the EDZ might represent a pathway for brine and could thereby facilitate brine inflow as well as radionuclide release from the repository. This is of particular importance for the EDZ around sealings with low permeability. Therefore, geometry, permeability and temporal development of EDZ are important parameters for performance assessment, especially during the period of compaction of the crushed salt until it reaches very low permeabilities to take over the barrier function inhibiting brine flow (Figure 1).

The permeability of EDZ will be reduced when the stress returns to a homogeneous state, e.g. after creep of the rock onto engineered barrier systems as plugs or seals. The models used to describe the reduction of permeability of EDZ with time under high pressure are derived from laboratory experiments. In order to check these models for time frames beyond that of laboratory experiments the analogue site at the Asse was studied.

The permeability distribution and the extension of the excavated rock salt were measured at various test sites in the Asse salt mine located in Northern Germany near Braunschweig (Bechthold, et al., 2004; Rothfuchs, et al., 2003; Wieczorek, et al., 2001). One drift, the so-called bulkhead drift, mined in 1911, is especially interesting with regard to the potential long-term behaviour of the EDZ. A 25 m long section of the drift was equipped with a liner of cast steel tubings in 1914, and the void between the liner and the drift surface was backfilled with concrete. This drift can be regarded as an industrial analogue for the development of an EDZ in a drift around a sealing. From the stress and permeability measurements taken in the EDZ around this bulkhead, information on the healing process of the EDZ in the long-term can be derived.

Figure 2 shows the permeability of the EDZ around the bulkhead drift compared to an open, unlined drift. Below the open drift, a typical EDZ is present. It extends about 1.5 m into the rock, and the permeability rises above 10<sup>-16</sup> m<sup>2</sup>. This confirms the results of a great number of permeability measurements at other test sites in the Asse salt mine. At all test sites with open drifts, an EDZ extension about 1.5 m into the floor and not more than 0.5 m into the walls was observed. Tests using various set-ups for measurements close to the open surface yielded permeability increases up to values from  $10^{-16}\,\mathrm{m}^2$  to  $10^{-15}$  m<sup>2</sup>, in comparison to around  $10^{-21}$  m<sup>2</sup> of the undisturbed salt. Around the lined part of the drift the permeability is completely different. Apart from the horizontal borehole close to the drift surface, all permeabilities are less than 10<sup>-19</sup> m<sup>2</sup> and thus considerably lower than the typical EDZ values. These lower permeabilities are due to the stress state with high normal and negligible deviatoric stress components, which is consistent with the results of supporting calculations. The original permeability of undisturbed salt, however, is not yet attained. Microstructural investigations on cores from both the lined and the unlined part of the drift seem to indicate that this may be due to the fact that the existing microfractures were closed by stress-induced plastic deformation, but did not completely disappear. In conclusion the conditions of natural dry rock salt in the Asse mine with about 0.02 wt.% water, 90 years under high compressive stress and negligible deviatoric stress were not sufficient to completely heal the EDZ around the bulkhead drift. However, this study clearly shows that a partial healing of the EDZ with permeability reduction of more than three orders of magnitude was observed.

Permeability
10<sup>-21</sup> m<sup>2</sup> 10<sup>-19</sup> m<sup>2</sup> 10<sup>-18</sup> m<sup>2</sup> 10<sup>-17</sup> m<sup>2</sup> 10<sup>-16</sup> m<sup>2</sup>

Figure 2: Measured permeability values in the boreholes around the lined drift (left) and below the open drift (right) in the Asse mine (Wieczorek, et al., 2001)

## Long-term stability of sealing materials: cements as example

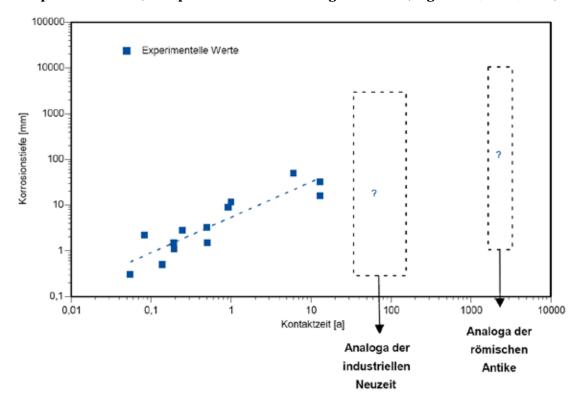
The most critical components of a sealing construction with respect to long-term stability are the materials for the sealing components. Here, concretes, clay materials (bentonites) and basaltic gravel are intended to be used (Bollingerfehr, et al., 2011, 2012). The long-term behaviour of these materials should be ensured by additional observations from analogues. As an example, cementitious materials are discussed in the following.

The application of cementitious materials in a repository for high-level waste in rock salt is preferably discussed with respect to sealing elements in shafts or drifts. A sealing has already been constructed in the repository for low- and intermediate-level waste in

Morsleben. As illustrated in Figure 1 the shaft and drift seals need to be tight until the salt grid backfill has been compacted down to a permeability, which is similar to that of the surrounding undisturbed rock salt (Bollingerfehr, et al., 2008). Consequently, beside the long-term evolution of the EDZ the long-term stability of the respective materials is of high importance.

Degradation of concrete can occur by interaction with fluids in the repository area. Laboratory experiments show that salt concrete is stable against highly mineralised sodium chloride solutions but unstable in the presence of magnesium-bearing solutions. Information about corrosion rates can be derived from laboratory experiments. In Figure 3 the results from corrosion experiments for concrete in highly mineralised magnesium sulphate solutions are shown. Most investigations are limited to a time frame of less than one year. Investigations performed over longer time frames, as those from the Asse mine (Kienzler, et al., 1999), are rare. Although experimental data suggest a linear correlation in double logarithmic scale, a prediction to time scales of 10 000 years is not possible, since no data for times later than 13 years exist. Here, results from analogue studies can deliver valuable information.

Figure 3: Experimentally determined corrosion depths of sulphate-resistant mortar and concretes with magnesium sulphate rich brines (dotted line: least square fit of the experimental data) and potential value of analogue studies (Hagemann, et al., 2009)



Roy, et al. (1982, 1983), Mallinson, et al. (1987) and Jull, et al. (1990) investigated the corrosion of historical concrete buildings in order to derive statements on the long-term durability of cementitious materials in repository projects. The samples under investigation were exclusively material from freshwater or atmospheric systems, i.e. not comparable to the conditions in a repository in rock salt. For this purpose, investigations on concrete buildings, which were in the past exposed to at least partly salinar conditions, like sea water, are necessary.

However, from archaeological-historical publications of Roman harbour buildings in the Mediterranean area some general statements can be derived (McCann, et al., 1987; Lamprecht, 1996). Important examples are the facilities in Pozzuoli, Cosa and Ostia in Italy or in Ampurias in Spain. Here, moles are still exposed to water of the Mediterranean, as at the time they were constructed 2 100-2 200 years ago. The buildings there have not only been exposed to sulphate and magnesium from the sea water but also to mechanical abrasion by wave breaking and disturbance by small animals and sea shells. All facilities are strongly damaged but still show a remarkable strength (16-20 N/mm²) after that long exposure time. Therefore, it can be assumed that a complete corrosion has not yet taken place. The mole in Ampurias consists of a concrete core, which is shielded by rocks. This core is still in a good shape.

The use of cement for construction purposes since 5000 B.C. has been proofed (Lamprecht, 1996). From lime mortar for floor pavement the application developed to mortar for walls finally leading to the so-called "Roman concrete". Still, many buildings from this time are giving evidence of this technological progress, which had not been reached again before the  $19^{th}$ - $20^{th}$  century.

From these investigations it can be deduced that the mechanical abrasion is the crucial damaging factor for the historical harbour buildings in contact with sea water. For advanced statements, especially on the corrosion progress at remaining buildings, detailed investigations under defined phyisco-chemical conditions are necessary. Also missing are explanations as to why some of the buildings are still existing and are in quite good shape and others are extremely damaged ruins.

Qualitative information is also of interest. In Wakely, et al. (1996) observations from sulphate-rich technical cement structures have been compiled to underpin results from laboratory experiments. An important aspect is that these structures show sharp corrosion fronts, which slowly enter the concrete. Behind the front the original chemical and physical properties of the cement exist, whereas the strength and hydraulic properties of the cement ahead of the front is significantly changed due to dissolution/precipitation processes. Similarly sharp fronts are simulated within the safety assessment for the low- and intermediate-level waste repository Morsleben. The analogue therewith supports the applied model in this aspect.

### **Compaction of crushed salt**

As discussed above the safety concept is based on low permeable rock salt backfill, which becomes responsible for the safety function "containment of the waste" after a distinct time. It is expected that for this compaction degree a time frame of several thousand years is needed. After repository closure, due to the surrounding rock pressure the residual void volume in the repository will be reduced by creep, and the backfill will be compacted. As a consequence of this compaction the porosity of the backfill material will be reduced, leading to a reduction of permeability and an increase in the backfill pressure. The increased backfill pressure results in a slowdown of the convergence process and the decreased permeability of the backfill causing a reduction of potential fluid flow in the repository area.

As mentioned, these rock mechanical processes occur over a long time scale which cannot fully be covered by laboratory experiments. In this context in situ experiments and natural analogue studies can play an important role to qualify and test the models used in PA and in backing-up the parameters derived from laboratory experiments. In particular, the investigation of old backfill material, which was used in abandoned salt mines and which has already been compacted for several decades, can provide suitable information on the long-term compaction process.

One study, which is presented here and can only be regarded as a pilot project was initiated several years ago (Brenner, et al., 1999). It was performed to identify the requirements for natural backfill samples and to find suitable sites in Germany, where

representative backfill samples can be found and investigated. As a first step, the most important requirements for an object to be investigated from a geotechnical point of view have been compiled, namely that the material should be similar to the backfill material foreseen in the German concept for a repository in rock salt, i.e. rock salt with grain sizes < 60 mm, preferably emplaced without additional water. It is important that information about the initial state of the backfill, the history of the backfilled cavity and the geology of the surrounding area is available.

In addition to the formulation of the geotechnical requirements, model calculations for various properties and lay-outs of a site have been performed to find the most promising examples. The aim was to identify the conditions under which a significant reduction of the backfill porosity (to about 10%), compared to an initial value of about 40% after emplacement, can be expected. The depth of the site, the moisture content of the crushed salt backfill and the existence of supporting anhydrite layers within the rock salt are all important parameters with potential impacts on the rate of the compaction process. The results of the calculations show that in cases where supporting anhydrite layers are present, the convergence will be so slow that no significant compaction can be expected within the first few hundred years, i.e. such sites are not suitable. If no supporting anhydrite layers are present, backfill will be reduced to porosities below 10% after some decades, if the backfill area is at a depth of at least 800 m. Therefore, the search has been focused on backfilled areas at around 800 m depth or greater. Based on this information, comprehensive research into backfilled sites in Germany and their condition has been carried out. As a result, suitable investigation targets in old salt mines at Sigmundshall, Riedel and Salzdetfurth, all in Northern Germany, have been identified.

The most suitable subjects were in Salzdetfurth and Riedel (see drill core in Figure 4) in Northern Germany. In both areas initial samples have been taken. These show that the material has – as expected – already been compacted. The backfilled areas at Sigmundshall were difficult to access and were therefore considered to be second in priority.



Figure 4: Final segments of a core drilled into old backfill from the 1 260/1 275 m floor of Riedel salt mine (Brenner, et al., 1999)

At this stage the pilot project ended. The results were promising and it is highly recommended to conduct further investigations to get additional data and to use this analogue for support of the models describing the compaction process of crushed salt in safety assessment.

## Role of industrial applications as analogues

A few geotechnical analogues have been discussed here. However, compared to the integrity and barrier function of the geological formation, not much analogue information is available for geotechnical barriers. In order to facilitate the use of analogues for this aspect of the safety case an objective of this workshop was to start a dialogue with industry and discussing and evaluating their experiences. It is the first time scientists from the oil and gas industry as well as from hazardous waste disposal have been invited for analogue discussion regarding rock salt. From the viewpoint of the safety case the following issues are of interest:

- In industry long-term experience exists with respect to the technology used for the construction of shafts and drifts. Information from industry might be helpful for optimisation of methods and concepts.
- In the mining industry and for disposal of hazardous waste selected open voids of the underground facilities have already been backfilled, for example for stabilisation of the mine, or are intended to be backfilled in the near future. Experiences here might also help to optimise methods, but differences in materials and their properties need carefully to be checked.
- In the oil and gas industry as well as in hazardous waste disposal shaft and drift sealings have been constructed, in order to effectively close the storage facility to avoid gas loss or to shield against intruding fluids. A thorough inspection of the existing facilities supports the demonstration that a drift or shaft can technically be realised, which must be done according to German requirements for disposal of heat-generating radioactive waste. Experiences might flow into construction projects or can be used as additional arguments for the safety case. This regards for example materials for sealing elements like bentonite or concrete, as well as gravel, as counter bearing materials.
- As shown by Lukas (Annex 1) industry has carried out research projects for shaft sealings and already constructed such sealings in repositories for hazardous waste.
   In addition Crotogino (Annex 2) demonstrated the tightness of sealings for gas storage caverns. From observation and investigation of both, information of the behaviour of such sealings under real conditions, particularly in the long-term can be derived and used in the safety case.
- In many old mines from industry backfilled underground areas exist. As already discussed, detailed investigations with respect to porosity, permeability and mineralogical structure of the backfill material, particularly of material which was emplaced several decades or even hundreds of years ago and was exposed to compaction, can support the models used for long-term evolution of porosity and permeability of the backfill due to convergence processes in the rock salt.

Several of these issues have been addressed in the presentations from Lukas (2013) and Crotogino (2013). The transparencies of both presentations are compiled in Annexes 1 and 2. The discussion with industry needs to be continued and the experiences and available information for each issue have to be further evaluated.

### References

- Bechthold, W., et al. (2004), Backfilling and Sealing of Underground Repositories for Radioactive Waste in Salt (BAMBUS-II Project), EUR 20621 EN, Luxembourg.
- Bollingerfehr, W., et al. (2011), Be-richt zum Arbeitspaket 5, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-272, ISBN 978-3-939355-48-9, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Köln.
- Bollingerfehr, W., et al. (2012), Endlagerauslegung und -optimierung. Bericht zum Arbeitspaket 6, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-281, ISBN 978-3-939355-57-1, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Köln.
- Bollingerfehr, W., et al. (2008), ISIBEL Abschlussbericht "Überprüfung und Bewertung des Instrumentariums für eine sicherheitliche Bewertung von Endlagern für HAW", Aufgabenpaket AP 1.2 Konzeptionelle Endlagerplanung, DBE Technology GmbH, TEC-20-2008-AP, Peine.
- Brenner, J., et al. (1999), Untersuchung von Altversatz als Analogon zur Konvergenz und Kompaktierung versetzter untertägiger Hohlräume im Salz über lange Zeiträume Phase I, GRS-Bericht 147, Braunschweig.
- Hagemann, S., M. Xie and H-J. Herbert (2009), Unsicherheits- und Sensi-tivitätsanalyse zur Korrosion von Salzbeton durch salinare Lösungen, Projekt Morsleben PSP-Element 9M 232 100 51, GRS-A-3458, Braunschweig.
- Herbert, H.-J., et al. (2004), "Alteration of Non-Metallic Barriers and Evolution of Solution Chemistry in Salt Formations in Germany", Proceedings of the Workshop on Engineered Barrier Systems (EBS) in the Context of the Entire Safety Case, Las Vegas, NV, USA, 14-17 September 2004, OECD/NEA, Paris.
- Jull, S.P. and T.P. Lees (1990), Studies of Historic Concrete, Final Report, EUR 12972 EN, Office for Official Publications of the European Communities, Luxembourg.
- Kienzler, B., et al. (2000), "Long-Term Leaching Experiments of Full-Scale Cemented Waste Forms: Experimental and Modelling Results", Nuclear Technology, Vol. 129.
- Lamprecht, H-O. (1996), Opus Caementitium Bautechnik der Römer, Beton-Verlag.
- Mallinson, L.G. and I.L. Davies (1987), A Historical Examination of Concrete, Final Report, EUR 10937 EN, Office for Official Publications of the European Communities, Luxembourg.
- McCann, A.M., et al. (1987), The Roman Port and Fishery of Cosa, Princeton University Press, Princeton, NJ.
- Mönig, J., et al. (2012), Sicherheits- und Nachweiskonzept. Bericht zum Arbeitspaket 4, Vorläufige Sicherheitsanalyse für den Standort Gorleben, GRS-277, ISBN: 978-3-939355-53-3, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH: Köln.
- Rothfuchs, T., et al. (2003), "Lessons Learned in Salt", European Commission CLUSTER Conference on the Impact of EDZ on the Performance of Radioactive Waste Geological Repositories, Luxembourg, 3-5 November 2003.

- Roy, D.M. and C.A. Langton (1982), Longevity of Borehole and Shaft Sealing Materials: Characterization of Cement-Based Ancient Building Materials, Technical Report ONWI-202, Office of Nuclear Waste Isolation, Columbus.
- Roy, D.M. and C.A. Langton (1983), Characterization of Cement-Based Ancient Building Materials in Support of Repository Seal Materials Studies, Technical Report BMI/ONWI-523, Office of Nuclear Waste Isolation, Columbus.
- Wakeley, L.D., T.S. Poole and J.P. Burkes (1994), Durability of Concrete Materials in High-Magnesium Brine, Sandia Report 93-7073.
- Wieczorek, K., P. Schwarzianeck and T. Rothfuchs (2001), "Self-Healing of Excavation-Disturbed Rocks in the Near Field of Underground Cavities Exemplary Measurements in Rock Salt and Interpretation of Preliminary Results", Proceedings of the Eurosafe Conference, Paris, 5-6 November 2001.

## NEA/RWM/R(2013)10

## Annex 1: Salt mining and hazardous waste disposal

#### V. Lukas

## The K+S Group - facts and figures

A world-wide leader in the sectors of:

- · special and standard fertilisers;
- · salt products;
- · more than 14 300 employees;
- revenue 2011: EUR 5.2 billion;
- listed on the DAX (Germany's most important stock index) since September 2008.

Figure 1: K+S group sites throughout the world



Recyclig

Residence

Recyclig

Recyc

Figure 2: K+S corporate structure

#### K+S Group profile

- · A K+S group business segment since 1991.
- · Disposal activities are centrally managed from Kassel.
- · Internationally active, with representatives in eight countries.
- · Pioneer of underground waste disposal; first facility world wide, Herfa-Neurode (1972).
- · Revenue 2011: EUR 87.7 million.
- · Europe's leading company in the field of underground waste disposal.

The K+S Group has two underground waste disposal facilities in Germany, Zielitz and Herfa-Neurode. They are indicated on the map in Figure 3.

Figure 3: K+S disposal facilities in Germany

1 – Zielitz, 2 – Herfa-Neurode



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Figure 4: Werk Werra - geology

Deposit thickness up to 500 metres | Gastight

Figure 5: Plan view of the room and pillar mining system

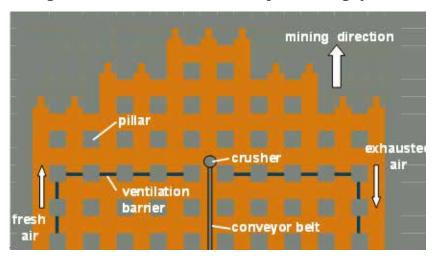
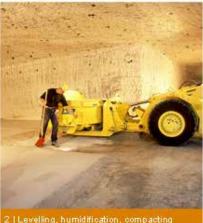


Figure 6: Mining - cavity preparation



Figure 7: Mining - roadway construction





## **UTD Herfa-Neurode**

## Multi-barrier system

Natural barriers:

- · salt (gastight) 300 m;
- · clay (watertight) 100 m;
- · bunter stone 500 m.

#### Artificial barriers:

- · waste packaging;
- · brick walls;
- · field dams;
- · disposal field dams;
- · watertight sealing of the shafts.

Figure 8: Installation at UTD Herfa-Neurode



#### Acceptance criteria

Wastes that are not acceptable for underground disposal include those that are:

- · radioactive;
- · liquid;
- · infectious;
- causing odour nuisance;
- · biodegradable;
- · increasing in volume;
- · releasing gases (toxic, self-ignitable, explosive);
- · explosive, highly or easily flammable;
- · insufficient stability for geomechanical conditions.

Figure 9: UTD Herfa-Neurode carefully conducts acceptance control, shaft transport, underground transport, storage chambers, sealing with walls









Figure 9: UTD Herfa-Neurode carefully conducts acceptance control, shaft transport, underground transport, storage chambers, sealing with walls (cont.)

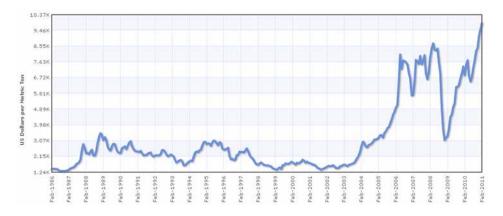


Figure 10: Sample storage at UTD Herfa-Neurode

Storage at this facility is characterised by internal identification codes and clear, traceable documentation – over 79 500 samples are stored



Figure 121 Copper price development in \$/t



Source: London Metal Exchange.

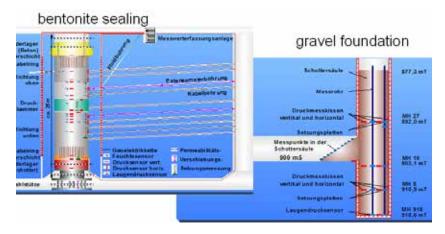


Figure 12: Transport of transformers to recycling facility

Figure 13: Products of cleaned transformers



Figure 14: Research project



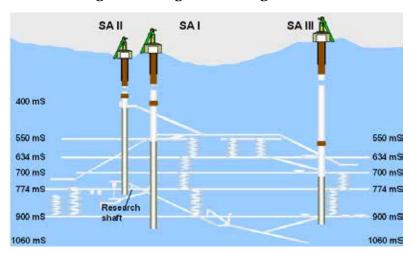


Figure 15: Long-term sealing of shafts

Figure 16: Concept for sealing the SA shafts

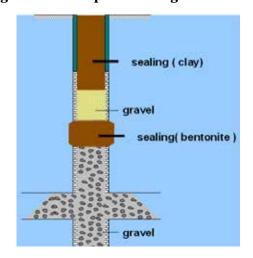
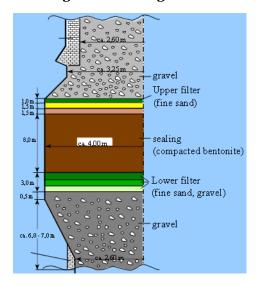


Figure 17: Sealing in salt



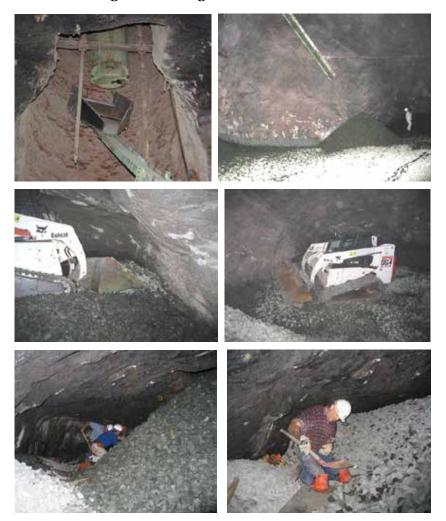
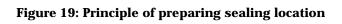


Figure 18: Filling of 400 mS, shaft SA II



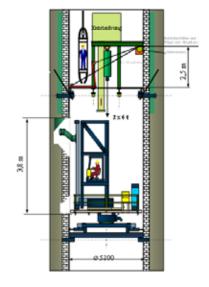


Figure 20: Cutting machine and cutter head





Figure 21: Bentonite pellets and granulate

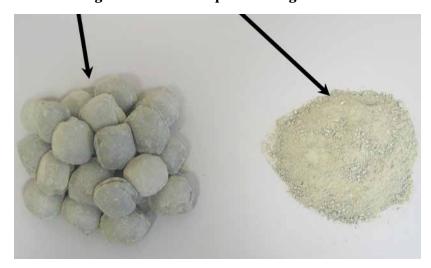


Figure 22: Production of bentonite pellets



Figure 23: Transport of bentonite



Figure 24: Building of bentonite seal



## NEA/RWM/R(2013)10

## Annex 2: Experiences from gas storage in salt caverns

#### F. Crotogino

KKB Underground Technologies GmbH, Hannover, Germany

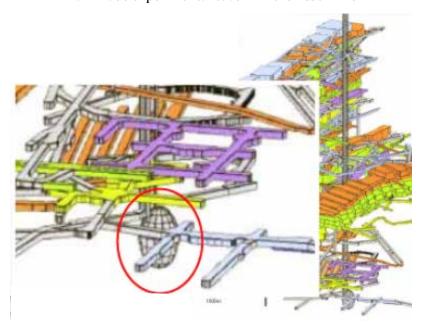
#### Introduction

#### Rad waste disposal (RWD) disposal in salt and salt cavern storage (SCSt)

In the past the SCSt industry benefited from RWD research, e.g. in the field of rock mechanics; in the future, the RWD community may benefit from activity in the SCSt industry, e.g. in the field of geological exploration.

Figure 1: The first idea - disposal of rad waste in salt caverns

Man-made experimental cavern in the Asse mine



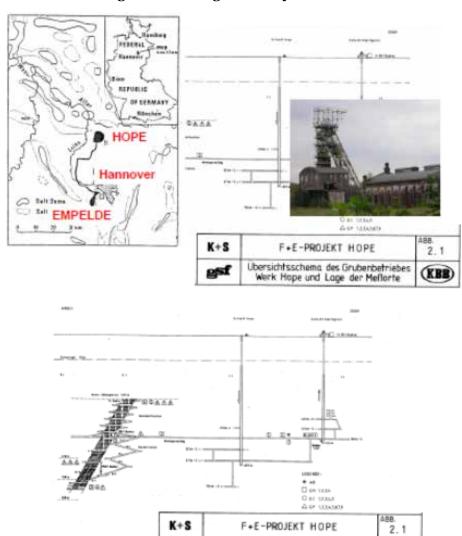


Figure 2: Flooding of the Hope salt mine

Figure 3: A video was produced about the flooding of the Hope mine

Übersichtsschema des Grubenbetriebes Werk Hope und Lage der Meßorte



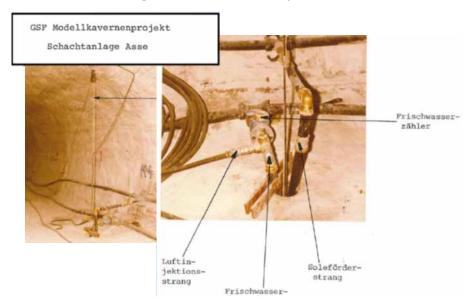


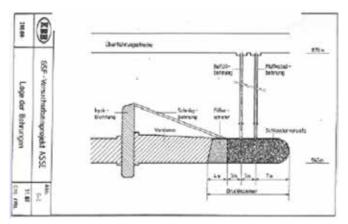
Figure 4: KBB consultancy for GSF

Figure 5: Brine from cavern leaking in Asse mine



Figure 6: GSF - experimental bulk head project Asse

Design of gas pressure chamber for permeability measurements



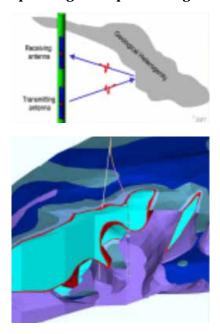
# Gas cavern technology benefits from radwaste research in Germany and the United States

## Geology

Technologies resulting from radwaste research in Germany and the United States which have benefited gas cavern technology include:

- · ground penetrating radar;
- · 3-D modelling of salt dome.

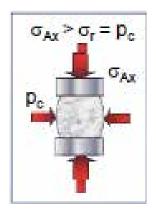
Figure 7: Examples of the outputs of ground penetrating radar (GPR) and 3-D modelling

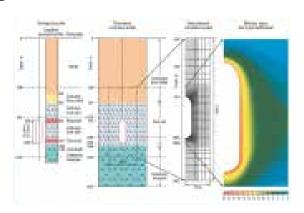


#### **Rock mechanics**

- · Material laws for rock salt.
- · Rock salt material parameters.
- · Numerical modelling.

Figure 8: Rock mechanics

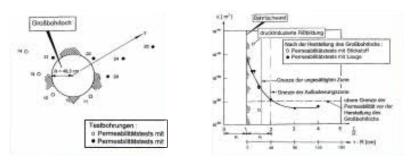




## Permeability of rock salt around borehole

- · In situ tests.
- · Lab tests.

Figure 9: Permeability of rock salt around borehole



## Radwaste disposal technology may benefit R&D for gas caverns

## Natural gas storage in Germany

Natural gas storage in Germany is characterised by:

- capacity of > 20% of annual consumption;
- about 200 salt caverns;
- · extremely high safety standards;
- $\cdot \hspace{0.1in}$  large know-how within the industry, academia and regulatory authorities.



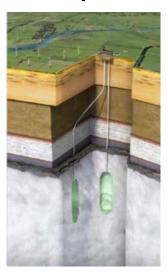
Figure 10: Natural gas storage in Germany

#### Recent key R&D issues in gas cavern engineering

Gas cavern engineering R&D issues with regard to geology include:

- ground-penetrating radar (GPR);
- · 3-D modelling;
- transition from homogeneous to inhomogeneous salt formations.

Figure 11: An example of 3-D modelling



## **Ground penetrating radar (GPR)**

Reflecting features within rock salt may represent:

- · layers of anhydrite;
- potassic seams;
- · layers of clay (stone);
- · interfaces between different salt units.

Receiving antenna

Transmitting antenna

Figure 12: How GPR functions

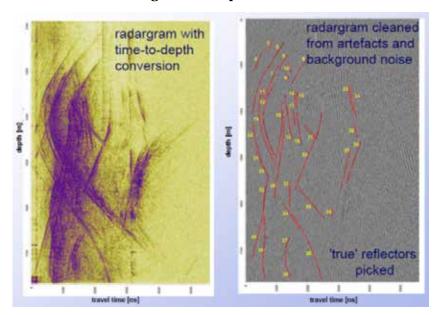
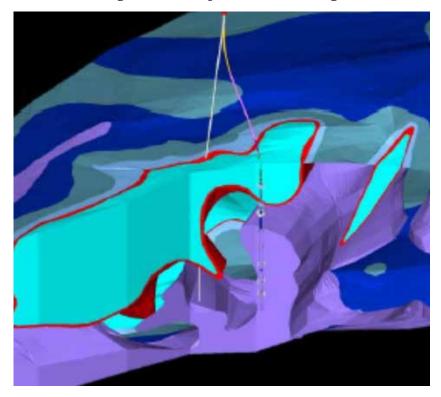


Figure 13: Outputs of GPR





Homogeneous case

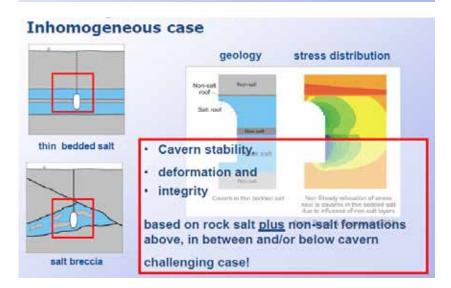
Geology Stress distribution

thick bedded salt

Cavern stability, and have bedded salt

deformation and
integrity
based on rock salt only: simple case!

Figure 15: Transition from homogeneous to inhomogeneous surrounding areas around cavern



#### **Rock mechanics**

- $\cdot \quad Load \ under \ frequent \ cycling/thermomechanical \ stress.$
- · Subsidence above cavern fields.

Figure 16: New challenge - frequency cycling

Seasonal versus multicycling storage

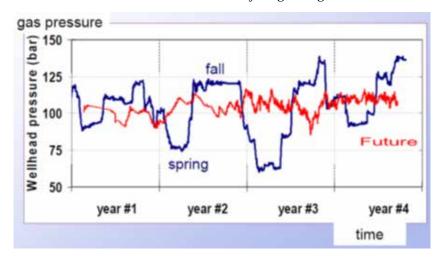
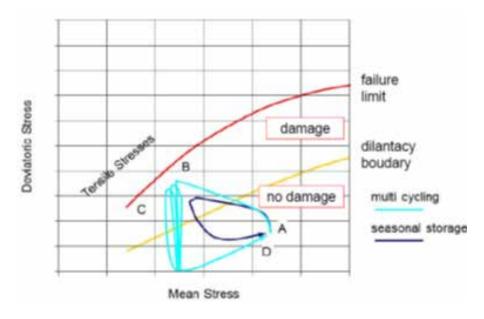


Figure 17: Stress path depending on gas cavern operational modus



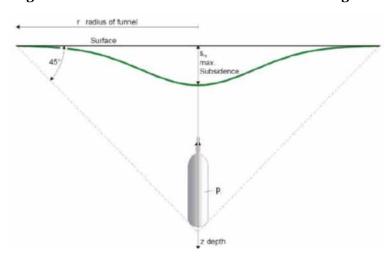
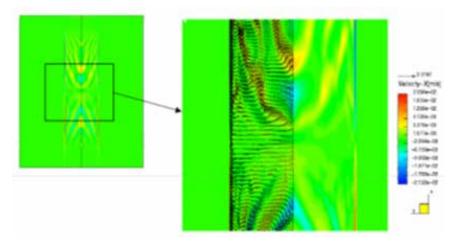


Figure 18: Surface subsidence due to cavern convergence

## Thermodynamics and fluid dynamics; computational fluid dynamics (CFD)

- Thermodynamic modelling of caverns assumes isotropic conditions: temperature <sup>1</sup> f(location).
- · CFD modelling allows computation of temperature = f(location, time).
- Applications:
  - proof of gas cavern tightness via mass balance;
  - accurate determination of stock in place;
  - determination of local water saturation.

Figure 19: Flow velocity distribution at steady-state conditions in a gas cavern



Source: Bannach, ESK: Köckritz, TU Freiberg; Kneer, TiNNiT.

Figure 20: Cavern abandonment (brine-filled)

Pressures in cavern vs. depth

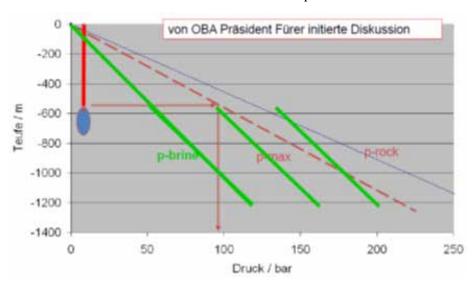
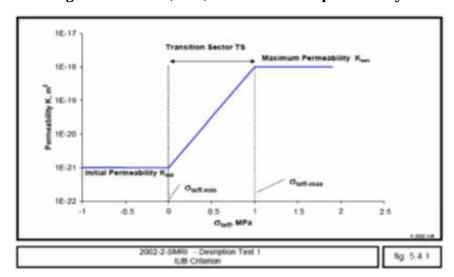


Figure 21: Fokker (Shell) - increase of salt permeability



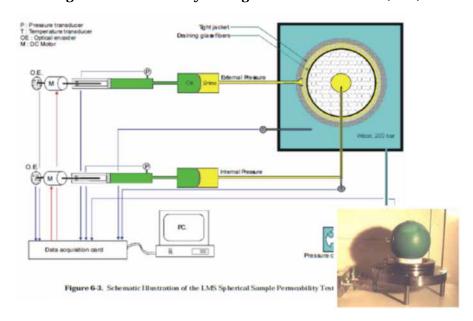
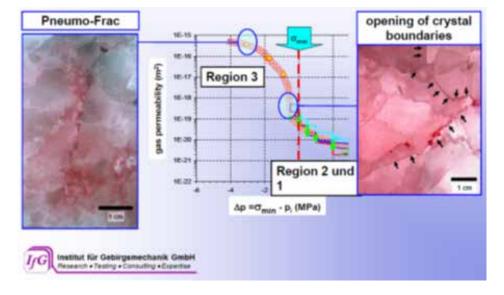


Figure 22: Permeability testing of cavern in salt ball (LMS)

Figure 23: Research on mechanism of brine penetration



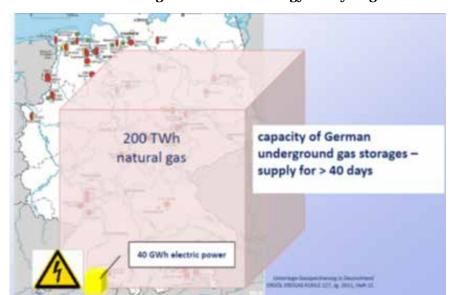
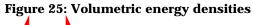


Figure 24: Gas-2-Power - storage of renewable energy via hydrogen in salt caverns



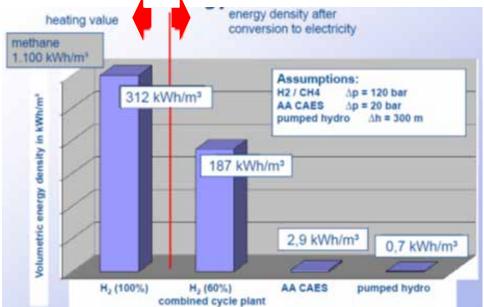




Figure 26: Capacity comparison

Figure 27: Demonstration projects for Power-2-Gas



## Session V

**Analogues for Chemical and Microbial Processes** 

## NEA/RWM/R(2013)10

# Ra-Ba precipitation in laboratory and large-scale evaporitic systems

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 $^{226}\mbox{Ra}$  is one of the radionuclides which is considered to be critical in long-term safety assessments for various radioactive waste repositories. Moreover the fate of radium is of high relevance for the environmental safety of scales produced in geothermal energy facilities, oil and gas exploration, and evaporation ponds for the management of residual solutions of uranium and other metals mining, coal mining and brackish water treatment plants. During the last decades the precipitation of (Ra,Ba)SO\_4(s) solid solution was extensively studied in laboratory experiments in dilute systems as well as in highly saline systems. The results of such laboratory scale experiments often serve in numerical safety assessments simulations. Large-scale evaporitic systems are used as man-made analogues to test the application of laboratory results on formation of (Ra,Ba)SO\_4(s) to radium retention processes in the geosphere.

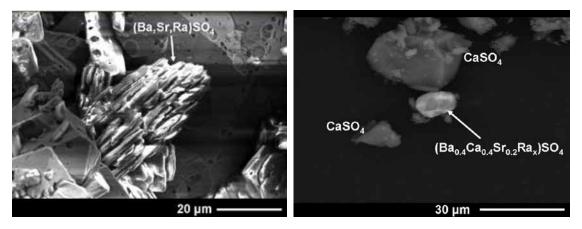
#### Introduction

## Relevance of $^{226}$ Ra to long-term safety assessments for radioactive waste disposal systems

In various studies on the long-term safety assessment of high-level waste disposal systems, e.g. in Japan, Sweden, Switzerland, and of low-level waste disposal systems in Germany, it is concluded that <sup>226</sup>Ra would be released as one of the dose dominating radionuclides from the repositories after 10 000s of years under certain conditions. For instance the safety assessment SR-Can (SKB, 2008) states that 226Ra is one of the main contributors to radiological dose in some of the studied scenarios. According to the recent safety assessment SR-Site, <sup>226</sup>Ra is simulated to be the dominant dose contributor after about 100 000 years (Wikberg, 2012). The <sup>226</sup>Ra dose depends directly on the aqueous concentration of radium in the near-field of the radioactive waste. In geochemical modelling studies, which are underlying SR-Can, SR-Site and other safety assessment studies, solubilities of pure solid Ra phases [mainly RaSO<sub>4</sub>(s)] are taken into account to calculate the aqueous Ra concentration in the near-field, while the formation of mixed solid phases are not considered (Berner, 1992; Berner and Curti, 2002; Curti, et al., 2010; Grandia, et al., 2008; Grivé, et al., 2013; Metz, et al., 2003). However, abundant information from early radiochemical experimental studies and recent studies on natural systems as well as man-made systems indicates that Ra readily forms solid solutions with barite. It is expected that the formation of (Ra,Ba)SO<sub>4</sub>(s) solid solutions (Figure 1) reduces the maximal aqueous Ra concentration by several orders of magnitude in comparison to the solubility with respect to a pure RaSO<sub>4</sub>(s) endmember (Grandia, et al., 2008; Prieto, et al., 2013). In repository systems for high-level waste significant amounts of stable barium

Figure 1: SEM images of ternary and quaternary Ra-bearing barite solid solutions

Left:  $(Ba,Sr,Ra)SO_4(s)$  crystal in a precipitate sampled from a geothermal energy plant in Southern Germany; right:  $(Ba,Ca,Sr,Ra)SO_4(s)$  crystal precipitated in Pond #1 of the K'tziot desalination facility, Southern Israel (see text)



isotopes will be present in spent nuclear fuel and vitrified radioactive waste. Ba isotopes represent the most abundant fission products in spent nuclear fuels of light water reactors and will occur in high level waste in concentrations of some grams per kg waste. Considering low-/intermediate-level waste repositories in rock salt diapirs in Germany, Ba is sufficiently abundant in the waste products and the anticipated saline solutions are rich in sulphate, thus significant amounts of barite could precipitate in the near-field. Based on these geochemical boundary conditions Ra incorporation into barite (radiobarite formation, respectively) is considered as a relevant process for an effective retention of <sup>226</sup>Ra.

As a pre-requisite of the quantitative modelling of  $^{226}$ Ra behaviour in a radioactive waste repository in rock salt, the uptake of radium by barite must be known quantitatively under repository-relevant conditions, i.e. in systems containing concentrated salt solutions. While the behaviour of dissolved Ra under low ionic strength environments is relatively well characterised (Porcelli and Swarzenski, 2003) and modelled (Agüero, 2005), Ra retention under saline conditions is hardly discussed in the literature (Kudryavskii and Rakhimova, 2007; Langmuir and Melchior, 1985; Paige, et al., 1998). The thermodynamics and kinetics of (Ra,Ba)SO<sub>4</sub>(s) formation were studied mainly in laboratory scale experiments though the closest approach to the natural environment is a deduction of empirical relationships from field scale studies. Recently, Rosenberg, et al. (2011a; 2011b; 2013) studied the (Ba,Ra)SO<sub>4</sub>(s) co-precipitation in laboratory and in large-scale evaporitic systems at ionic strength (I) ranging from 0.7 to 7.0 mol·(kg  $H_2O$ )<sup>-1</sup>. The present communication focuses on the retention of Ra by barite at ambient temperature studied in these recent large scale field experiments and laboratory scale evaporation batch experiments.

#### Discussion

#### Formation of (Ba,Ra)SO<sub>4</sub>(s) solid solutions

Retention of radium by  $(Ba,Ra)SO_4(s)$  may occur via: i) coprecipitation; ii) reprecipitation ("cocrystallisation") processes:

$$x \times Ra_{aa}^{2+} + (1 - x) \times Ba_{aa}^{2+} + SO_{4,aa}^{2-} = (Ra_x Ba_{1-x})SO_4(s)$$
 (1)

$$x \times Ra_{aa}^{2+} + BaSO_4(s) = (Ra_x Ba_{1-x})SO_4(s) + x \times Ba_{aa}^{2+}$$
 (2)

In their classical study on  $(Ba,Ra)SO_4(s)$  coprecipitation Doerner and Hoskins (1925) derived an analytical solution to calculate the concentration-based apparent partition coefficient,  $K_{\Phi,barite}$ :

$$K'_{D} = \frac{\ln(Ra_{t}^{2+}/Ra_{0}^{2+})_{aq}}{\ln(Ba_{t}^{2+}/Ba_{0}^{2+})_{aq}}$$

where  $Ra_i$  and  $Ba_i$  are quantities (mol) of  $Ra^{2+}$  and  $Ba^{2+}$  in solution, respectively, and the subscripts t and 0 refer to time = t and 0, respectively. The values for K¢ reported for Ra retention by barite are usually in the range 1.0 to 2.0 (Rosenberg, 2012; Rosenberg, et al., 2011b and references therein). Doerner and Hoskins (1925) considered the value of 1.8 as representing the thermodynamic value because it was evaluated from very slow coprecipitation experiments.

If radiobarite is formed by coprecipitation from an aqueous solution containing Ba, Ra and sulphate, the concept of solid solution/aqueous solution equilibria as well as the relevant activities of aqueous species and species in the solid are straightforward to apply. Activities of aqueous species in saline solutions are accurately described by the Pitzer model (Pitzer, 1973; 1979). Paige, et al. (1998) determined Pitzer parameters for the RaSO<sub>4</sub>-H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O system. Pitzer parameters for the highly important RaCl<sub>2</sub> interaction were estimated by Rosenberg, et al. (2011a).

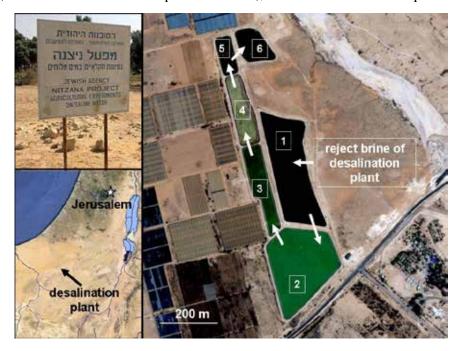
In case of aqueous Ra contacting a pre-existing barite, there will be exchange reactions occurring at the barite surface. The overall reaction involves dissolution of the barite and the consecutive reactions of the released  $Ba^{2+}$  and sulphate ions with the dissolved  $Ra^{2+}$  to form radiobarite. In this case the achievement of a "true equilibrium" requires the complete dissolution of the initial barite and the subsequent reprecipitation of sulphate,  $Ba^{2+}$  with  $Ra^{2+}$  to produce a solid solution phase with a homogenous composition (Prieto, et al., 2013). With respect to natural systems and repository conditions, it is less clear whether formation of a  $(Ba,Ra)SO_4(s)$  solid solution will occur via dissolution and reprecipitation of the complete barite crystals or will be the solid solution process occur only in the surface regions.

# $(Ba,\!Ra)SO_4(s)$ coprecipitation in laboratory evaporation experiments and in a field scale evaporation system

In laboratory batch experiments, Rosenberg, et al. (2011b) studied (Ba,Ra)SO<sub>4</sub>(s) coprecipitation under varying ionic strength conditions by evaporating a fresh reject brine sampled from the K'tziot desalination plant (near Nitzana in the Israeli Negev desert; Figure 2). The NaCl rich brines were supersaturated with respect to gypsum, celestite and barite. During evaporation the ionic strength increased from 0.7 mol·(kg H<sub>2</sub>O)<sup>-1</sup> up to halite saturation and gypsum and barite precipitation commenced within a few days. Although barite precipitation was quantitatively minor compared to that of gypsum, its role in determining the removal of  $^{226}Ra^{2+}$  was considerable. The observed linear relationship between precipitated  $^{226}Ra^{2+}$  and  $Ba^{2+}$ , as opposed to the non-concurrent precipitation of <sup>226</sup>Ra<sup>2+</sup> both with Ca<sup>2+</sup> and Sr<sup>2+</sup> indicated that barite was the main host mineral phase for Ra<sup>2+</sup>. In these laboratory experiments, Rosenberg, et al. (2011b) determined a concentration-based apparent partition coefficient of  $K\phi_{\text{,barite}} = 1.04 \pm 0.01$ . They demonstrated that the difference between their low value of K. b., barite compared to Kø,barite of Doerner and Hoskins (1925) was mainly the result of a kinetic effect but was also slightly affected by the ionic strength. Taking into account the RaCl<sub>2</sub> Pitzer parameters estimated by Rosenberg, et al. (2011a), activities of Ra<sup>2+</sup> and Ba<sup>2+</sup> in solution were calculated in order to determine an activity-based partition coefficient, which accounts for the ionic strength effect. Furthermore, they determined a rational activity coefficient of the RaSO<sub>4</sub> component in the radiobarite, g<sub>kaSO4</sub> = 1.1, corresponding to a Guggenheim (Redlich-Kistler) interaction parameter of  $a_0 \sim 0.1$ .

Figure 2: Evaporation ponds of the K'tziot desalination plant (Israel)

Satellite images of the north-western Negev desert and the six sequential evaporation ponds near Nitzana (downloaded from www.maps-for-free.com); arrows indicate the flow path of the brine



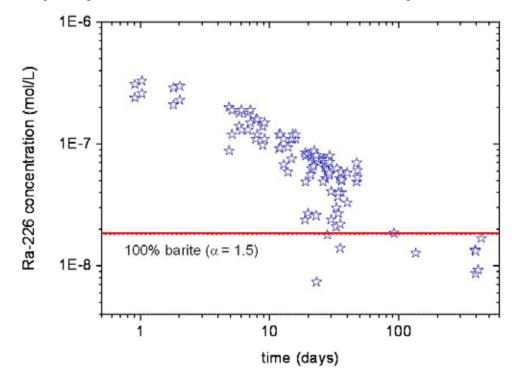
In agreement with the laboratory evaporation experiments, chemical analysis of solid samples collected from the K'tziot evaporation ponds (Figure 2) revealed that in the field study the precipitation of Ra is also concurrent with Ba, indicating the formation of Ra bearing solid solution. Despite the complexity of the K'tziot evaporation system – a super saturated system containing various solutes and various solid phases at high ionic strength – it was demonstrated that Ra was removed through the precipitation of a (Ra,Ba)SO<sub>4</sub>(s) solid solution; either as a binary solid solution, or as a part of a ternary (Ra,Ba,Sr)SO<sub>4</sub>(s) solid solution with compositional zoning with zones that are Ba-rich and zones that are Sr-rich. The results of the field scale evaporation study were comparable to those of the laboratory-based experiments, suggesting that in the complex field system, as in the laboratory, the same factors affect the formation of the radiobarite solid solution.

#### (Ba,Ra)SO<sub>4</sub>(s) reprecipitation in laboratory evaporation at low ionic strength

Curti, et al. (2010) investigated the uptake of  $^{226}Ra^{2+}$  by barite by means of reprecipitation experiments in pure  $H_2O$ , 0.1 mmol·(kg  $H_2O$ )<sup>-1</sup> Na<sub>2</sub>SO<sub>4</sub> solution and 0.01 mol·(kg  $H_2O$ )<sup>-1</sup> NaCl solution. Unfortunately, the studied radiobarite/solution systems did not reach steady state even in their longest experiment (120 days). The range of  $K\phi$ <sub>,barite</sub> reported by Curti, et al. (2010) is relatively low ( $K\phi$ <sub>,barite</sub> = 0.12 to 0.61). Using barite reprecipitation batch experiments in 0.1 mol·(kg  $H_2O$ )<sup>-1</sup> NaCl solution, Bosbach, et al. (2010) showed that the aqueous Ra concentration is controlled by the solubility of a (Ba,Ra)SO<sub>4</sub>(s) solid solution and many orders of magnitude below the  $^{226}Ra^{2+}$  concentration, which is controlled by RaSO<sub>4</sub>(s). In both studies the formation of the (Ba,Ra)SO<sub>4</sub>(s) solid solution was described by a regular model. Curti, et al. (2010) derived from their experimental data an interaction parameter  $a_0$  in the range of 1.5 to 2.5, whereas in the study of Bosbach, et al. (2010) an  $a_0$  = 1.5 was derived under the assumption that the whole barite fraction in their experiments were equilibrated with Ra (Figure 3).

## Figure 3: Removal of aqueous $^{226}$ Ra $^{2+}$ in a barite/ 0.1 mol·(kg H<sub>2</sub>O) $^{-1}$ NaCl suspension as a function of time

After doping the barite/NaCl solution with  $^{226}$ Ra<sup>2+</sup>, the  $^{226}$ Ra<sup>2+</sup> concentration decreases continuously until approaching a steady state after more than 100 days (data taken from Bosbach, et al., 2010). The line indicates the calculated  $^{226}$ Ra<sup>2+</sup> concentration corresponding to a complete equilibration (100%) of a radiobarite with an interaction parameter of  $a_0 = 1.5$ .



#### **Concluding remarks**

The complementary laboratory and large scale evaporation studies under saline conditions (Rosenberg, 2012; Rosenberg, et al., 2011a; 2011b; 2013) demonstrate that (Ba,Ra)SO<sub>4</sub>(s) coprecipitation is affected by the ionic strength of the solution and the kinetic of the solid solution precipitation. As a result of these two effects the radiobarite composition in the evaporation systems is less Ra-enriched compared to the equilibrium radiobarite composition determined in dilute systems by Doerner and Hoskins (1925). Still, published experimental data of Ra/barite coprecipitation (e.g. Doerner and Hoskins, 1925; Gordon, 1955; Gordon and Rowley, 1957; Rosenberg, et al., 2011b; 2013) show concordantly that the partition coefficient is larger from, or equal to, unity, implying a relative enrichment of Ra in the solid phase and Ra<sup>2+</sup> depletion in the aqueous solution. In contrast, partition coefficients significantly smaller than unity are derived from reprecipitation (cocrystallisation) studies of Curti, et al. (2010) and Bosbach, et al. (2010), which imply a relative enrichment of Ra<sup>2+</sup> in the solution and not in the solid phase. Since there is an apparent discrepancy between coprecipitation experiments and reprecipitation experiments, it is questionable whether this discrepancy is a result of kinetic effects which are not yet recognised, or whether the discrepancy is based on a misinterpretation of any of the experimental data (Rosenberg, 2012). Further studies are needed to examine to what extent the radiobarite co- and reprecipitation systems approach near-equilibrium conditions. Prieto, et al. (2013) emphasised that - amongst other aqueous solution-solid solution systems - Ra<sup>2+</sup>/Ba<sup>2+</sup>/sulphate/(Ba,Ra)SO<sub>4</sub>(s) systems may need a long time to approach "true equilibrium".

With respect to safety assessments of nuclear waste disposal, the usage of data of coprecipitation studies and those of reprecipitation studies will end up with significantly different estimations of Ra solubility (see discussion in Rosenberg, 2012). As incorporation of Ra into pre-existing is considered a relevant scenario for Ra retention (Curti, et al., 2010), the relatively low Ra partition coefficients derived from the reprecipitation experiments may be used for safety assessments, as long as a discrepancy between the different Ra retention phenomena exist.

#### References

- Agüero, A. (2005), "Performance Assessment Model Development and Parameter Acquisition for Analysis of the Transport of Natural Radionuclides in a Mediterranean Watershed", Science of the Total Environment, 348, 32-50.
- Berner, U. (1992), Radionuclide Concentration Limits in the Near-Field of a Repository for Spent Fuel and Vitrified High-Level Waste, Project Opalinus Clay, Technical Report 02-10, NAGRA, Wettingen, Switzerland.
- Berner, U. and E. Curti (2002), Radium Solubilities from SF/HLW Wastes Using Solid Solution and Coprecipitation Models, Paul Scherrer Institute, Villigen, Switzerland, Internal Report TM-44-02-04.
- Bosbach, D., M. Böttle and V. Metz (2010), Experimental Study on Ra<sup>2+</sup> Uptake by Barite (BaSO<sub>4</sub>). Kinetics of Solid Solution Formation via BaSO<sub>4</sub> Dissolution and Ra(x)Ba(1-x)SO<sub>4</sub> (Re)Precipitation, SKB TR-10-43, Svensk Kärnbränslehantering AB, Stockholm.
- Curti, E., et al., (2010), "Radium Uptake During Barite Recrystallization at  $23 \pm 2^{\circ}$ C as a Function of Solution Composition: an Experimental <sup>133</sup>Ba and <sup>226</sup>Ra Tracer Study", Geochimica et Cosmochimica Acta, 74, 3553-3570.
- Doerner, H.A. and W.M. Hoskins (1925), "Coprecipitation of Radium and Barium Sulfates", Journal of the American Chemical Society, 47, 662-675.
- Gordon, L. (1955), "Slow Precipitation Processes Application of Precipitation from Homogeneous Solution to Liquid-Solid Distribution Studies", Analytical Chemistry, 27, 1704-1707.
- Gordon, L. and K. Rowley (1957), "Coprecipitation of Radium and Barium Sulfate", Analytical Chemistry, 29, 34-37.
- Grandia, F., J. Merino and J. Bruno (2008), Assessment of the Radium-Barium Coprecipitation and its Potential Influence on the Solubility of Ra in the Near-Field, SKB TR-08-07, Svensk Kärnbränslehantering AB, Stockholm.
- Grivé, M., et al., (2013), Handling of Uncertainty in Thermodynamic Data, Report to Svensk Kärnbränslehantering AB, SKBdoc id 1396560, Amphos 21, Barcelona, Spain, 30 pp.
- Kudryavskii, Y. and O. Rakhimova (2007), "Coprecipitation of Radium with Barium Sulfate from Salt Solutions", Radiochemistry, 49, 541-544.
- Langmuir, D. and D. Melchior (1985), "The Geochemistry of Ca, Sr, Ba and Ra Sulfates in Some Deep Brines from the Palo Duro Basin, Texas", Geochimica et Cosmochimica Acta, 49, 2423-2432.

- Metz, V., W. Schüßler and B. Kienzler (2003), Experimentelles Programm zur Bestätigung der Ergebnisse von standortspezifischen Modellrechnungen für das FB Asse: Abschluss- bericht Teil 7, Geochemische Modellrechnungen für die Schachtanlage Asse, FZK-INE 011/03, Forschungszentrum Karlsruhe, Germany, 97 pp.
- Paige, C.R., et al. (1998), "Solution Equilibria for Uranium Ore Processing: The BaSO<sub>4</sub>-H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O System and the RaSO<sub>4</sub>-H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O System", Geochimica et Cosmochimica Acta, 62, 15-23.
- Pitzer, K.S. (1979), "Theory: Ion Interaction Approach", in R.M. Pytkowicz (Ed.), Activity Coefficients in Electrolyte Solutions, CRC Press, Boca Raton, Florida, USA.
- Pitzer, K.S. and G. Mayorga (1973), "Thermodynamics of Electrolytes. II. Activity and Osmotic Coefficients for Strong Electrolytes with One or Both Ions Univalent", The Journal of Physical Chemistry, 77, 2300-2308.
- Porcelli, D. and P.W. Swarzenski (2003), "The Behavior of U- and Th-Series Nuclides in Groundwater", in J.J. Rosso and H.P. Ribbe (Eds.), Uranium Series Geochemistry, 317-361.
- Prieto, M., J.M. Astilleros and L. Fernández-Díaz (2013), "Environmental Remediation by Crystallization of Solid Solutions", Elements, 9, 195-201.
- Rosenberg, Y.O. (2012), The Fate of Radium in Evaporitic Systems, PhD Thesis, Ben-Gurion University of the Negev.
- Rosenberg, Y.O., V. Metz and J. Ganor (2011a), "Coprecipitation of Radium in High Ionic Strength Systems: 1. Thermodynamic Properties of the Na-Ra-Cl-SO<sub>4</sub>-H<sub>2</sub>O System Estimating Pitzer Parameters for RaCl<sub>2</sub>", Geochimica et Cosmochimica Acta, 75, 5389-5402.
- Rosenberg, Y.O., et al. (2011b), "Coprecipitation of Radium in High Ionic Strength Systems: 2. Kinetic and Ionic Strength Effects", Geochimica et Cosmochimica Acta, 75, 5403-5422.
- Rosenberg, Y.O., V. Metz and J. Ganor (2013), "Ra-Ba Coprecipitation in a Large Scale Evaporitic System", Geochimica et Cosmochimica Acta, 103, 121-137.
- SKB (2008), Long-Term Safety for KBS-3 Repositories at Forsmark and Laxemar A First Evaluation. Main Report of the SR-Can Project" SKB TR-06-09, Svensk Kärnbränslehantering AB.
- Wikberg, P. (2012), "The Outcome of Scientific Research and its Implementation in the Swedish Nuclear Waste Disposal Programme", ATW Atomwirtschaft Atomtechnik, 57, 102-105.

# Consideration of hydrocarbons at the Gorleben salt dome in a preliminary safety assessment

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The occurrence of hydrocarbons in rock salt at the Gorleben site raised questions pertaining to its suitability for final disposal of heat-generating radioactive waste. A preliminary safety assessment investigated the impact of hydrocarbons on the safety assessment, identified uncertainties, needs for research, development and exploration. The geomechanical and geochemical impact of hydrocarbons was studied. Criteria and consequences for design of the repository were discussed. Conclusions on the safety concept, disposal concept and repository design were derived. The presently known concentration and distribution of hydrocarbons do not exclude a suitability of the Gorleben site for disposal.

#### **Discussion**

#### Hydrocarbons in salt rock and in Gorleben

Hydrocarbons (HC) are a mixture of different compounds from the elements carbon and hydrogen. They are generated as intermediate products from degradation of biomass. Depending on size and mass HC can be liquids or solids. Due to their low density they tend to migrate upwards through rock layers. A HC deposit can be formed if tight rock layers hinder the upward migration and trap HC.

Since rock salt is impermeable some shale oil and gas can also be trapped in salt rocks. HC within salt rocks are well known and found, if fracture networks are generated due to tectonic stress, if the hydraulic pressure exceeds the lithostatic pressure or if HC are generated in situ from incorporated organic masses at deposition. The origin of HC can be concluded from its geochemical signature.

HC can be intracrystalline in salt minerals or intercrystalline on grain boundaries of salt minerals within mm-scales. Intercrystalline inclusions dominate in general. HC can be released due to mining and subsequent damage to the rock fabric.

Gases (including HC) represent a hazard to mining due to:

- · blowout of rock salt and gas;
- generation of explosive gas/air mixtures;
- pressure impact of sudden gas flows;
- chokedamp and toxic effect of gases.

Releases of such magnitudes are not known for HC in salt domes. Safety-relevant effects are mainly related to operational safety due to slow release of burnable gases. Preventive measures are mandatory when releases are expected or possible.

There are only a few site-specific publications concerning HC at the Gorleben site (Popp, et al., 2002; Gerling and Faber, 2001; Fischer, 2000; Bornemann, et al., 2008). During the exploration HC were met locally as spots in the galleries (Figure 1).

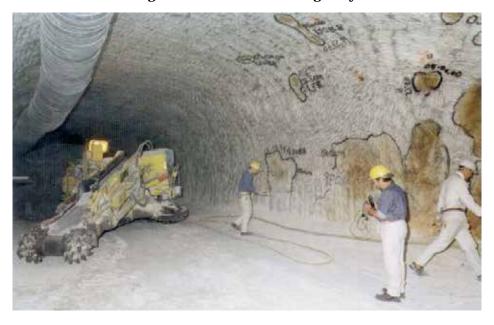


Figure 1: Release of HC in a gallery

Source: Photo courtesy of DBETec.

According to (Weber, et al., 2011) the HC in the Gorleben salt dome occur mainly within the Knäuelsalz (z2HS1) as nodules (see Figure 2). Gaseous compounds range from 0.3 to 20 g/Mg. The methane content is even lower. The condensed HC ranges from 40 to  $250 \text{ ml/m}^3$ . The overall content is estimated to be below 0.1% and is localised in spots of  $1 \text{ m}^3$ . The HC are autochthonous products from the organic matter of the underlying Stassfurt carbonate (z2SK).

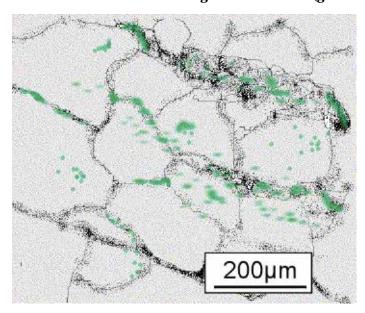


Figure 2: Distribution of HC within grain boundaries (green = HC)

## Are HC a concern for long-term safety?

HC can represent a safety hazard as a flammable gas in the operational phase. This is not considered here.

In the post-operational phase of the repository the HC are considered to compromise possibly barrier integrity by some processes. The processes are:

- · combustion;
- · radiolysis;
- pyrolysis;
- · chemical reactions;
- · thermochemical reaction of sulphates.

Combustion is not relevant in the post-operational phase since oxygen is not available. The low concentration of HC and oxygen in a closed repository does not favour combustion.

Radiolysis is only effective for HC in close vicinity of containers and considered to be low because of the shielding.

Thermal effects (pyrolysis) will overcompensate for any radiolytical effects. Any decomposition rate of HC may increase due to the heat-generating radioactive waste.

The chemical reactions are mostly negligible, e.g. halogenation of HW with iodine.

The thermochemical reduction of sulphate (TSR) is considered to be possibly relevant because of temperature and the amount of sulphate and HC available.

# Impact of HC on the system evolution

The safety concept of the repository design is based on containment of radionuclides by geotechnical and geological barriers. There may be some hydromechanical as well as geochemical impacts due to the HC. The thermal reduction of sulphates with HC and gas generation may change the rock volume. In addition, the degradation products of HC may change the corrosion behaviour of containers. Complexation may change the release and mobility of radionuclides.

#### **Technical studies**

Technical studies are summarised in Bracke, et al. (2012).

Assessment of HC in the vicinity of a POLLUX container

Within the preliminary safety assessment of the Gorleben site (VSG) a repository concept of disposal of POLLUX containers in drift was developed. Based on the size of the POLLUX container, drift and an excavation damaged zone (EDZ) of 2 m a rock volume of 571  $\rm m^3$  per container is impacted from which are 471  $\rm m^3$  EDZ [cf. Figure 3(a)]. It was assessed in Weber, et al. (2011) that every 10 m a nodule of 1  $\rm m^3$  could contain up to 1 l of HC [see Figure 3(b)]. The EDZ could allow HC and degradation products to migrate into the backfill of the gallery. HC beyond the EDZ are not considered since this seems to be arbitrary and not justified.

It is estimated that the EDZ will reach 2-3 spots with HC and that therefore approximately 3 l of HW will migrate into the pore volume of the backfill.

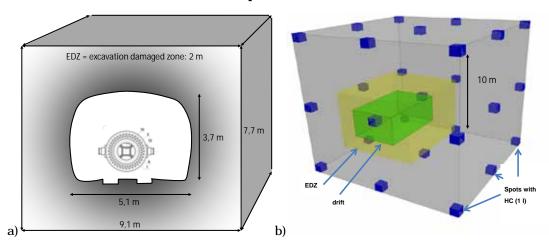


Figure 3: (a) Disposed POLLUX container, (b) rock volume with spots of HC around drift and EDZ

Thermal sulphate reduction (TSR) with HC

The TSR is a well-known process in geological formations explored for oil and gas. The TSR is known for temperatures above  $80\text{-}100^{\circ}\text{C}$  in geology. This process leads to oxidation of HC and formation of H<sub>2</sub>S and water:

$$CH_4 + CaSO_4 \otimes CaCO_3 + H_2S + H_2O$$

This reaction leads to changes in the specific rock density and rock features. The intermediate compounds of degradation of HC may alter the corrosion behaviour of containers and the complexation of radionuclides.

The amount of generated water is small compared to the water content of the EDZ and backfill. The overall potential generation of gas by TSR is small compared to the potential gas generation by corrosion of containers.

The impact of degradation products of HC by TSR on corrosion of containers and complexation of radionuclides is considered to be small due to the large amount of iron compared to the concentration of HC degradation products in dissolution.

Based on hydromechanical modelling any impact on geomechanical rock features does not start until a few vol.% of HC.

Thermal sulphate reduction (TSR) with hydrogen

A similar process to reduction of sulphates with HC is the reduction of sulphates with hydrogen. The corrosion of containers with water generates hydrogen:

$$Fe + 2 H_2O \otimes Fe(OH)_2 + H_2$$

This hydrogen may react with the anhydrite, which is an abundant component in rock salt:

$$4 H_2 + Ca^{2+} + SO_4^{2-} \otimes H_2S + 2 H_2O + Ca^{2+} + 2 OH^{-}$$

The hydrogen sulphide may react with iron oxides and release water:

$$Fe(OH)_2 + H_2S \otimes FeS + 2 H_2O$$

Although the redox processes do not balance each other, they may reduce the amount of generated hydrogen and hydrogen sulphide gas.

These reactions proceed until one of the educts is used up.

The stability field for coexistence of sulphides and carbonates is quite small in the Eh-pH diagram for all temperatures. The reactions will remain unchanged but may exhibit different rates.

Some studies with highly saline solutions containing sulphides up to 200 mg/l did not show any effect on the corrosion rate which could be contributed to sulphides.

#### Criteria

There was some demand to define criteria concerning the hydrocarbons and the Gorleben site. Criteria can be applied for the suitability/unsuitability of the site or for the design of the repository.

The site is suitable if the operational safety is guaranteed, the barrier integrity is given, the containment-providing rock zone is provided and other requirements are met. If one of these conditions is not met, then the site is not suitable.

The criteria for design are geology/geometry, physics/chemistry and mobility or migration of HC. Table 1 lists the criteria, parameter, the threshold/effect of the parameter and possible measures for compensation.

Criteria Parameter **Threshold** Effect Possible compensation Adjustment of utilisation Amount/ HC-content per volume Hydro-mechan. impact; (exclusion of areas with (small/large scale) if less than few % then negligible concentration Geologic/ unfavourable bounding geometric conditions) Distribution of HW/ Distribution Geochem. interaction with HC anhydrite in rock salt Change of design Safety distance between areas with HC and disposed T < 80°C TSR/pyrolysis negligible waste Physical/ Temperature and/or chemical Change of repository design TSR, release via EDZ, pyrolysis, T > 80°C (e.g. lowering temperature thermo-expansion requirement) Auxiliary ventilation Phase Gas/salt outbursts (gas/liquid/ solid) Safety distances Temporarily closed areas for exploration and Mobility/ Maximum content of Pore pressure Operational safety emplacement migration fluids Minimising impact of mining (reduction of EDZ) Geochemical interactions due to Fluid content for release of fluids (e.g. corrosion, Sealing of potentially gas generation TSR) problematic areas

Table 1: Criteria for design

# Conclusions

The most relevant process to consider with HC is the thermal sulphate reduction (TSR). Also the TSR with hydrogen may be relevant.

A significant impact on the release of radionuclides due to gas generation, corrosion or complexation is not expected for presently known concentrations of HC. The known content of HC and its extrapolation does not exclude the suitability of the Gorleben site as a repository.

Some quantitative criteria have to be assessed for a safety case for licensing which will concern the suitability of Gorleben and the design of the repository.

Subsequently, further development of models for long-term safety and consequences is required.

Some details on HC in Gorleben were studied. It was not possible to improve the models for long-term safety analysis to include these effects. Also, some experimental and theoretical work is to be done in order to obtain more data on TSR and its thermodynamics under conditions of a repository for heat-generating waste. Especially, investigation and modelling of fluid behaviour at grain boundaries, their impact on microstructural processes and on transport is required to assess the potential impact.

Thermodynamic modelling concerning balances of reactions and their kinetics is also required to assess the importance of the chemical reactions (corrosion, TSR with hydrogen) for long-term safety.

#### References

- Bornemann, O., et al. (2008), "Projekt Gorleben, Standortbeschreibung Gorleben. Teil 3: Ergebnisse der über- und untertägigen Erkundung des Salinars", Geologisches Jahrbuch, Vol. C, 73, 211 Seiten, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR), Hannover.
- Bracke, G., et al. (2012), Berücksichtigung der Kohlenwasserstoffvorkommen in Gorleben in der VSG, Bericht der Arbeitsgruppe Kohlenwasserstoffe, GRS-285, ISBN 978-3-939355-61-8.
- Fischer, M. (2000), "Gefügekundliche Untersuchung von Steinsalz im Hinblick auf die Verteilung und Bindungsform von Kohlenwasserstoffen und die Bestimmung des Volumenanteils der Kohlenwasserstoff-Einschlüsse", Diplomarbeit, 59 Seiten, Institut für Geowissenschaften, Kiel.
- Gerling, P. and E. Faber (2001), Projekt Gorleben, Geologische Bearbeitung der Erkundungssohle Dokumentation der chemischen Analysen von gasförmigen und flüssigen Kohlenwasserstoffen, 20 Seiten, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR), Hannover.
- Popp, T., M. Fischer and H. Kern (2002), "Gefügekundliche Untersuchungen zur Verteilung von Kohlenwasserstoffen (KW) im Steinsalz", Meyniana, Vol. 54, S. 131-154.
- Weber, J.R., J. Hammer and O. Schulze (2011), Empfehlungen der BGR zur Berücksichtigung der Kohlenwasserstoff-Vorkommen im Hauptsalz des Salzstockes Gorleben im Rahmen einer vorläufigen Sicherheitsanalyse, Projekt Gorleben GE442800000, Bundesanstalt für Geowissenschaften und Rohstoffe (BGR), Hannover.

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# Analogues for microbial effects in rock salt

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An overview of experimental findings and field studies on survival of microbes in ancient rock salt is given, which suggests that indigenous microbes can survive over geological periods of time in rock salt. Additionally, microbes introduced into rock salt from the surface as a result of contamination may remain viable there for a considerable time. Microbial species retaining the ability to actively metabolise in saturated brines include those which produce corrosive substances and gases. Therefore, microbial activity may be of relevance for long-term performance of deep geological repositories in rock salt and needs to be evaluated.

#### Introduction

The site and design of a repository should fulfil key post-closure safety requirements for final disposal of high-level radioactive waste and spent nuclear fuel in deep geological formations, such as: i) containing the waste; ii) isolating the waste from the biosphere; iii) attenuating or delaying migration of radionuclides to the biosphere. A demonstration that this is indeed the case is carried out by performance assessment.

An important stage of the performance assessment is the identification and documentation of the features, events and processes that may be relevant to long-term safety (Mazurek, et al., 2003). Amongst others, those related to microbial activity (if any) in rock salt need to be considered. Such a consideration requires understanding of whether microbes may be active in rock salt, and, if so, how and to what extent they can influence the long-term performance of a deep geological repository (DGR) built there.

The purpose of this work is to give an overview of experimental findings and field studies concerning the survival of microbes in ancient rock salt, which can be used as natural analogues for assessing the fulfilment of the key post-closure safety requirements for a DGR in rock salt. An evaluation of possible microbial effects on the long-term performance of a DGR in rock salt, on the contrary, is not within the scope of the present work and should be carried out in the follow-up studies.

#### Discussion

# Abundance of microbes in ancient rock salt

As late as 1981, mined rock salts were described as microbe-free by microbiologists. This perception of salt mines as sterile environments is still widespread, although there is no doubt any more that ancient halite deposits support substantial populations of halophilic bacteria and haloarchaea (Grant, 2004). For example, G-Seep brine from the underground workings of the United States Department of Energy Waste Isolation Pilot Plant (WIPP) contained  $7 \cdot 10^4$  to  $3 \cdot 10^6$  cells ml<sup>-1</sup> (Francis and Gillow, 1993), brines from

the Winsford and Boulby salt mines in the United Kingdom contained up to 4  $^{\circ}$  10<sup>7</sup> and 1.9  $^{\circ}$  10<sup>6</sup> cells ml<sup>-1</sup>, respectively (McGenity, et al., 2000). These microbial abundances are similar to those observed for deep claystone and other sediment formations (Meleshyn, 2011). In this regard it should be noted that numerical abundances of microbes in natural saline environments may have been substantially underestimated in the past due to populations of Nanohaloarchaea with a cell diameter of ~0.6 mm not being accounted for; they were discovered very recently and had eluded previous detection (Narasingarao, et al., 2012).

Salado formation hosting the WIPP was reported to contain viable and cultivable populations of halophilic microbes, which are scattered heterogeneously throughout the available mine areas with up to 10 000 colony forming units per ml found in G-Seep 1 brine and none found in Room 7 of the WIPP (Vreeland, et al., 1998). When considering these numbers, it should be added that, as is now widely accepted, about 99.9% of microbes from natural environments resist cultivation in the laboratory (Anderson, et al., 2006; Little and Lee, 2007).

Some of the populations mentioned above might have been non-indigenous to the salt formations and represent species introduced there due to human activity during the construction of the mine areas. However, the mere fact of their survival in the rock salt is of importance for the performance assessment studies of the final repositories for high-level radioactive waste and spent nuclear fuel in rock salt. Indeed, the non-indigenous microbial species will inevitably be introduced into the repository system as a result of repository excavation as well as placement of radioactive waste, backfill and sealing materials.

There are few negative controls in studies of ancient evaporites, e.g. no microbes were recovered from salts that had been subjected to burial temperatures of  $\sim 160^{\circ}$ C (Dombrowski, 1963). Similarly, no microbes were found in brine from a highly deformed potash seam within a salt diapir, which had previously been deeply buried and heated to at least  $80^{\circ}$ C (McGenity, et al., 2000).

# Viability of microbial populations in ancient halite

Living microbes were claimed to be isolated from rock salts as early as the 1960s – the primary Zechstein salt of about 250 million years old, and the Precambrian salt of about 650 million years old – using rigorous techniques to avoid contamination (Dombrowski, 1963). The isolated bacteria with optimum growth temperature of 45-55°C were embedded in the crystalline structure of the salt and not in the capillary crevices as evidenced by petrographic thin sections of the salt (Figure 1). The isolation of microbes from the primary Zechstein salt sampled at another location than in the latter study were reproduced using numerous controls and extensive precautions against surface contamination (Bibo, et al., 1983) and are currently considered credible, albeit not incontrovertible (McGenity, et al., 2000: Mormile, et al., 2003: Grant, 2004: Adamski, et al., 2006).

Extraction and cultivation of a 250-million-year-old halotolerant bacterium from a brine inclusion in a halite crystal from the WIPP site (Salado formation) were claimed in a more recent report (Vreeland, et al., 2000), which has received significant publicity because of the extreme sterilisation techniques used to avoid contamination by modern micro-organisms.

Critics questioned this claim by arguing that the large, single-crystal nature of the sampled halite is not typical of primary halite deposition, so that the fluid inclusion and the viable bacterium in them may represent much more recent features (Hazen and Roedder, 2001). However, the disputed fluid inclusions were found to contain evaporated Permian seawater (Satterfield, et al., 2005).

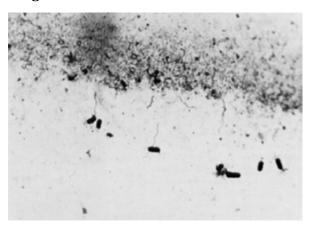


Figure 1: Microbes in the Precambrian salt

Source: Dombrowski, 1963.

Based on an evolutionary rate of 1.5 nucleotide substitutions per 100 bases in 16S rDNA per 50 million years and the observation that the 250-million-year-old bacterium from the WIPP site is 99% identical to a modern bacterium from the Dead Sea, critics argued that the extracted bacterium must be a modern one (Nickle, et al., 2002; Hebsgaard, et al., 2005). However, observations of extreme similarity of haloarchaea in 23-, 121- and 419-million-year-old salts from Spain, Brazil and the United States suggested that salt formations represent a natural gene bank for Earth's biosphere through dissolution and reprecipitation of salt depositions as a result of the active water cycle and tectonic processes (Park, et al., 2009).

Further critics were concerned with the issue of DNA stability over long periods of time and the lack of independent replication of the observation (Willerslev and Hebsgaard, 2005). However, the same authors later succeeded in evidencing DNA repair and bacterial survival for 0.5 million years in permafrost (Johnson, et al., 2008).

# Sources of energy for survival of microbes in halite

Metabolic activity – even a very low one – such as DNA repair requires an in situ energy source. This poses a further question on the mechanism of microbial survival in fluid inclusions. Microbes in fluid inclusions from Death Valley with an age of 12 up to 100 thousand years were often observed to be co-trapped with the Dunaliella algae (Figure 2), which contain up to 7 M glycerol,  $C_3H_5(OH)_3$ , in their cytoplasm and release it into brine (Schubert, et al., 2010). Besides, although virtually all hypersaline lakes contain large amounts of dead plant material, the Salado formation contains no sign of fossilised organics but detectable populations of cellulose-degrading microbes (Vreeland, et al., 1998). This points out that microbes may survive in rock salt over long periods of times by utilising co-deposited organics as energy and nutrient sources.

Furthermore, domal and bedded rock salt often contain high pressure pockets with gas inclusions, which have the potential to outburst (Ehgartner, et al., 1998). Gas inclusions in US domal salt are considered to be of primary origin (from decomposition of trapped organics during the original salt deposition) or of secondary origin (penetration of gases during the domal rise) (Kupfer, 1978). In the Gulf of Mexico Coast salt mines, they tend to align along a single, black, clay-bearing salt bed (Kupfer, 1978). US domal salts contain 90-99% CH<sub>4</sub>, whereas in Germany, CH<sub>4</sub> and CO<sub>2</sub> are frequently reported as the main components of gas mixtures (Ehgartner, et al., 1998). Outburst salt was reported to have an average gas content of up to 2 and 18 m³ per tonne of rock salt in the United States and Germany, respectively, whereas gas content in normal salt is <0.003 m³ t<sup>-1</sup> (Ehgartner, et al., 1998).

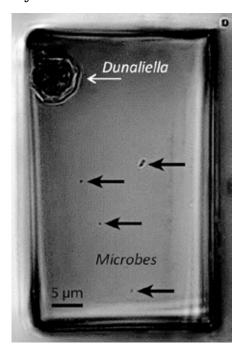


Figure 2: Microbes and Dunaliella algae co-trapped in a 12 000-year-old fluid inclusion from Death Valley

Source: Schubert, et al., 2010.

 $CO_2$  can be utilised as an electron acceptor by fermentative and methanogenic microbes, whereas anaerobic methanotrophic microbes gain energy by utilising  $CH_4$  as an electron donor in the process of methane oxidation (Meleshyn, 2011). In the latter process, either sulphate or Fe(III) is used as an electron acceptor. Sulphate is a major anion in halite brines with the concentrations of e.g. up to 340 mmol per kg  $H_2O$  observed for the fluid inclusions in halite of the Salado formation hosting the WIPP site (Lowenstein, et al., 2005). Fe(III) may be present in rock salt as Fe(III)-(oxyhydr)oxides, which give a characteristic orange colour to some Zechstein rock salt layers (Urai and Boland, 1985; Siemann and Ellendorf, 2001), or as clay (Kupfer, 1978; Siemann and Ellendorf, 2001; Swanson, et al., 2012).

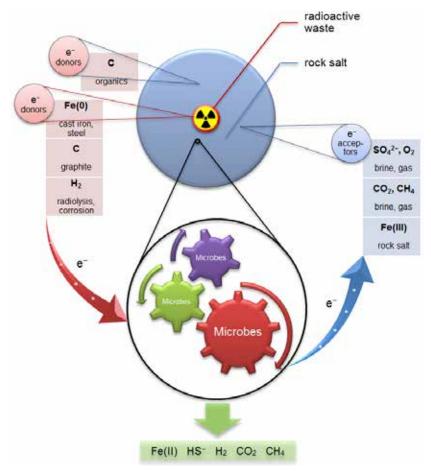
Furthermore, fluid inclusions within halite grains often contain hydrocarbons, which would suffice for microbial activity (McGenity, et al., 2000; Siemann and Ellendorff, 2001). Further organic carbon may be associated with carbonate clay facies in salts, as in the Bresse and Mulhouse basins in France containing 0.1-7% organic carbon (Pironon, et al., 1995). An additional energy source ( $H_2$ ) may result from gamma and beta irradiation (e.g. from  $^{40}$ K) of NaCl and formation of colloidal Na (Turkin, et al., 2006). Brine interaction with colloidal Na as a result of, e.g. grain boundary migration can then lead to a  $H_2$  release according to the reaction  $2Na + 2H_2O$  ®  $2NaOH + H_2$  (Schléder, et al., 2007). Possible sources of energy for microbial activity and the major corresponding products of microbial metabolism in a DGR in rock salt are presented in Figure 3.

# Survival of microbes in fluid inclusions in halite

In order for microbes to survive in a high-saline environment, an osmotic balance between the latter and microbial cytoplasm is required. One of the two survival strategies is to accumulate high concentrations of  $K^+$  (Oren, 2011) with intracellular KCl concentrations being as high as of 4-5 M (Grant, 2004). Hence, colloidal Na or clusters of Na atoms may form in the vicinity of microbes and provide a continuing supply of  $H_2$  to microbes as a result of reaction of metallic Na with water. Such a mechanism might

Figure 3: Schematic summary of electron donors and acceptors and microbial metabolism products in a DGR in rock salt

Microbes utilise redox processes to gain energy for metabolism and growth by directing the flow of electrons from a donor to an intermediate, intracellular acceptor and from that to a terminal, extracellular acceptor



explain, e.g. the observation of sylvite (KCl) along with CH<sub>4</sub> and organic matter within (pseudo)secondary fluid inclusions in blue halite in Zechstein salt from the Kłodawa mine in Poland (Wesełucha-Birczyńska, et al., 2008).<sup>9</sup>

Related to the intracellular accumulation of  $^{40}$ K is the question about the radiation resistance of microbes. Calculations suggest that a lethal dose would not be generated due to such an accumulation even over  $10^9$  years (Grant, 2004). Besides, e.g. Halobacterium species growing optimally in 4 M NaCl has been shown to resist to 5 000 Gy of  $^{60}$ Co gamma rays (Kottemann, et al., 2005). In another example, microbial biofilms developed on the walls of the damaged Three Mile Island reactor during its defueling operation in 1986 despite a radiation dose rate of 100 Gy  $h^{-1}$  (Wolfram, et al., 1997).

The issue of microbial incorporation and survival in fluid inclusions in halite was extensively investigated in laboratory settings with 40 strains of halophilic bacteria (Norton and Grant, 1988). All tested strains were observed to retain viability for a minimum of

<sup>9.</sup> The mechanism of formation of metallic Na in halite still being disputed as discussed by Sonnenfeld (1995) and Wesełucha-Birczyńska, et al. (2008), metallic Na is considered as the most common reason for the blue discoloration of halite (Sonnenfeld, 1995).

six months and motility for at least three weeks within fluid inclusions in laboratory-grown halite crystals. In another study, even non-halophilic bacteria (Pseudomonas aeruginosa, which were genetically modified to produce green fluorescent protein) might have remained viable for at least 13 months after being incorporated in fluid inclusions – and not in the crystal matrix – of laboratory-grown halite crystals (Figure 4) (Adamski, et al., 2006). A further study reported on an aseptic extraction (by a combined microscope microdrill/micropipette system) and cultivation of viable haloarchaea from 97 000-year-old fluid inclusions in primary halite crystals deposited in Death Valley (Mormile, et al., 2003).

<u>0 30</u> μm

Figure 4: Fluorescing cells of P. aeruginosa (seen as bright spots) in fluid inclusions in laboratory-grown halite crystals

Source: Adamski, et al., 2006.

# Microbial effect on local brine environment

Crystallisation of halite crystals was observed to begin 24-28 hours earlier in all samples containing bacteria than in sterile controls in the above-discussed experiments by Norton and Grant (1988). Similarly, crystallisation was observed in brines seeded with halophilic bacteria at a salinity of 225 g l<sup>-1</sup> NaCl (~100 g l<sup>-1</sup> NaCl below the saturation with halite), whereas no halite crystals formed in the unseeded controls (Castanier, et al., 1999). The latter study attributes this effect to the above-discussed intracellular accumulation of KCl and which tends to saturate or even oversaturate salt brine with respect to NaCl in the vicinity of microbes. The driving force for such a local increase of NaCl concentration would be the intracellular/extracellular concentration gradient, which is characterised by a ratio of 0.3-0.8 reported by Christian and Waltho (1962) for, e.g. NaCl extracellular concentration of 236 g l<sup>-1</sup>. It was further speculated by Castanier, et al. (1999) that such a process could have played a significant role in deposition of rock salt over geological times.

#### Solute constraints on microbial survival in rock salts

From a study of the 0.05-5.05 M MgCl $_2$  gradient of the seawater at the hypersaline brine interface of Discovery Basin – a large, stable brine lake located on the Mediterranean Sea floor and almost saturated with MgCl $_2$  – it has been concluded that the upper MgCl $_2$  concentration for microbial life is about 2.3 M. Above this concentration, biological macromolecules are suggested to be denatured by MgCl $_2$  (Hallsworth, et al., 2007). However, a conflicting evidence on microbial activity in Discovery brine and a primary reliance of the conclusion by Hallsworth, et al., (2007) on cultivation method leaves open the possibility of finding uncultivated extremophiles adapted to the existence in MgCl $_2$  brines (Sass, et al., 2008; Kminek, et al., 2010).

## Near-field microbial effects in performance assessment

The only available for a DGR in rock salt, WIPP conceptual model for microbiology considers production of CO<sub>2</sub> and biosorption to be significant microbial contributions to the performance of the repository and assumes that the produced CO<sub>2</sub> will come from denitrification (4%) and sulphate reduction (96%). Methanogenesis is not considered to contribute to biodegradation and gas production (Swanson, et al., 2012).

These assumptions are based on the observations that: i) sulphate-reducing bacteria can actively produce  $H_2S$  up to salt concentration of 350 g  $l^{-1}$  (Oren, 2011); ii) denitrification occurs up to similar salt concentrations (Oren, 2011); iii) methanogenesis by reduction of  $CO_2$  occurs only up to ~90 g  $l^{-1}$ , whereas methanogenesis by fermentation of methylated amines, dimethylsulfide or methanol occurs up to ~210-250 g  $l^{-1}$  (Oren, 2011).

These observations represent the current state of knowledge in microbiology of high-salt environments and should be considered generally applicable in the performance assessment of DGR in rock salt. One should bear in mind, however, that e.g. methane contents in fluid inclusions in halite layers without underlying oil and gas deposits can be as high as ~42%, as observed in the Zechstein rock salt in northern Poland (Kovalevych, et al., 2008). This indicates that microbial methane production might have occurred in the Zechstein halite during its post-depositional history [see also the discussion on the observation of sylvite and  $CH_4$  in (pseudo)secondary fluid inclusions above]. This further suggests that the issue of methanogenesis should be considered for incorporation in performance assessment models if substantiated by the geologic or geochemical realities of the rock salt in question to host a DGR.

#### Far-field microbial effects

According to Sassen, et al. (1994), formation of an anhydrite cap rock occurs at the first stage of dissolution of the underlying halite diapir by meteoric water. Carbonate cap rock then develops largely by downward replacement of anhydrite cap rock, which is sometimes separated from the anhydrite cap rock by a transitional zone of gypsum, elemental sulphur and some metal sulphides. The carbonate cap rock of Damon Mound salt dome in Texas with an estimated weight of about 33  $^{'}$  10 $^{6}$  tonnes has been found to originate from bacterial oxidation of an equivalent of about 5.4  $^{'}$  10 $^{6}$  m $^{3}$  of crude oil (Sassen, et al., 1994). A utilisation of CO<sub>2</sub>, crude oil,and CH<sub>4</sub> – in this preference order – by the sulphate-reducing bacteria is considered as the probable source of carbon in carbonate cap rocks (Posey and Kyle, 1988; Sassen, et al., 1994).

Biodegradation of hydrocarbons and microbial sulphate reduction have been found not only to be responsible for low-temperature formation of carbonate minerals but also for formation of elemental sulphur and metal sulphides in the cap rocks of the coast of the Gulf of Mexico (United States) and Balhoul formation (Tunisia) salt domes (Posey and Kyle, 1988; Bechtel, et al., 1996; Saunders and Thomas, 1996). These studies have revealed that sulphate supplied from anhydrite dissolution is reduced by sulphate-reducing bacteria to  $H_2S$  in amounts more than sufficient to rapidly precipitate convective supply of Fe, Zn and Pb from the sequences underlying the salt domes (Posey and Kyle, 1988; Bechtel, et al., 1996; Saunders and Thomas, 1996). Even at such a relatively high extent of  $H_2S$  production, the activity of the sulphate-reducing bacteria was still below the production capacity of their population due to a limited supply of sulphate (and not of hydrocarbons) despite its high availability in close proximity to salt domes (Bechtel, et al., 1998).

# **Conclusions**

Microbes capable of exerting impact, either negative or positive, on the long-term performance of a radioactive waste repository are indigenous to rock salt and, possibly

added by microbial species introduced during mining or drilling activities, may be present in abundances comparable to those observed in deep subsurface sediments.

Rock salts can contain electron donors and acceptors in amounts sufficient for microbes to remain active for very long periods of time. Additional sources of electron donors and acceptors will inevitably be added to the repository system as a result of repository excavation and placement of radioactive waste, backfill and sealing materials.

Hence, a microbiological exploration of repository environments in rock salts and an evaluation of the maximum microbial effect in long-term performance assessments for a deep geological repository for high-level radioactive waste and spent nuclear fuel in rock salt appear to be necessary. In case the microbial effect might be assessed to be negative and significant, an elaboration of possible measures to inhibit or impede microbial activity might be an option for a repository design.

#### References

- Adamski, J.C., J.A. Roberts and R.H. Goldstein (2006), "Entrapment of Bacteria in Fluid Inclusions in Laboratory-Grown Halite", Astrobiology, 6, pp. 552-562.
- Anderson, C.R., et al. (2006), "In Situ Ecological Development of a Bacteriogenic Iron Oxide-Producing Microbial Community from a Subsurface Granitic Rock Environment", Geobiology, 4, pp. 29-42.
- Bechtel, A., et al. (1996), "Biodegradation of Hydrocarbons and Biogeochemical Sulfur Cycling in the Salt Dome Environment: Inferences from Sulfur Isotope and Organic Geochemical Investigations of the Bahloul Formation at the Bou Grine Zn/Pb Ore Deposit, Tunisia", Geochimica et Cosmochimica Acta, 60, pp. 2833-2855.
- Bechtel, A., M. Pervaz and W. Püttmann (1998), "Role of Organic Matter and Sulphate-Reducing Bacteria for Metal Sulphide Precipitation in the Bahloul Formation at the Bou Grine Zn/Pb Deposit (Tunisia)", Chemical Geology, 144, pp. 1-21.
- Bibo, F-J., R. Söngen and R.E. Fresenius (1983), "Vermehrungsfähige Mikroorganismen in Steinsalz aus primären Lagerstätten", Kali u Steinsalz, 8, pp. 367-373.
- Castanier, S., et al. (1999), "The Salt Ooids of Berre Salt Works (Bouches du Rhône, France): The Role of Bacteria in Salt Crystallization", Sedimentary Geology, 125, pp. 9-21.
- Christian, J.H.B. and J.A. Waltho (1962), "Solute Concentrations Within Cells of Halophilic and Nonhalophilic Bacteria", Biochimica et Biophysica Acta, 65, 506-508.
- Dombrowski, H. (1963), "Bacteria from Paleozoic Salt Deposits", Annals of New York Academy of Sciences, 108, pp. 453-460.
- Ehgartner, B., J. Neal and T. Hinkebein (1998), Gas Releases from Salt, SAND98-1354, Sandia National Laboratories, Albuquerque, NM.
- Francis, A.J. and J.B. Gillow (1993), "Effects of Microbial Processes on Gas Generation Under Expected WIPP Repository Conditions", Annual Report Through 1992, BNL-49518, Brookhaven National Laboratory, Upton, NY.
- Grant, W.D. (2004), "Life at Low Water Activity", Philosophical Transactions of the Royal Society London B, 359, pp. 1249-1267.

- Hallsworth, J.E., et al. (2007), "Limits of Life in MgCl<sub>2</sub>-Containing Environments: Chaotropicity Defines the Window", Environmental Microbiology, 9, pp. 801-813.
- Hazen, R.M. and E. Roedder (2001), "How Old are Bacteria from the Permian Age?", Nature, 411, p. 155.
- Hebsgaard, M.B., M.J. Phillips and E. Willerslev (2005), "Geologically Ancient DNA: Fact or Artefact?", Trends in Microbiology, 13, pp. 212-220.
- Johnson, S.S., et al. (2007), "Ancient Bacteria Show Evidence of DNA Repair", Proc. of the National Academy of Sciences of the United States of America, 104, pp. 14401-14405.
- Kminek, G., et al. (2010), "Report of the COSPAR Mars Special Regions Colloquium", Advances in Space Research, 46, pp. 811-829.
- Kottemann, M., et al. (2005), "Physiological Responses of the Halophilic Archaeon Halobacterium Sp. Strain NRC1 to Desiccation and Gamma Irradiation", Extremophiles, 9, pp. 219-227.
- Kovalevych, V.M., et al. (2008), "Geochemical Aureoles Around Oil and Gas Accumulations in the Zechstein (Upper Permian) of Poland: Analysis of Fluid Inclusions in Halite and Bitumens in Rock Salt", Journal of Petroleum Geology, 31, pp. 245-262.
- Kupfer, D.H. (1978), "Problems Associated with Anomalous Zones in Louisiana Salt Stocks, USA", Proceedings of the 5<sup>th</sup> International Symposium on Salt, The Northern Ohio Geological Society, Inc., Cleveland, Ohio, pp. 119-134.
- Little, B.J. and J.S. Lee (2007), Microbiologically Influenced Corrosion, John Wiley & Sons, Inc., Hoboken, New Jersey.
- Lowenstein, T.K., et al. (2005), "The Major-Ion Composition of Permian Seawater", Geochimica et Cosmochimica Acta, 69, pp. 1701-1719.
- Mazurek, M., et al. (2003), Features, Events and Processes Evaluation Catalogue for Argillaceous Media, Organisation for Economic Co-operation and Development/ Nuclear Energy Agency, NEA No. 4437, OECD/NEA, Paris.
- McGenity, T.J., et al. (2000), "Origins of Halophilic Microorganisms in Ancient Salt Deposits", Environmental Microbiology, 2, pp. 243-250.
- Meleshyn, A. (2011), Microbial Processes Relevant for Long-Term Performance of Radioactive Waste Repositories in Clays, Report GRS-291, BMWi-FKZ 02 E 10548, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Braunschweig.
- Mormile, M.R., et al. (2003), "Isolation of Halobacterium Salinarum Retrieved Directly from Halite Brine Inclusions", Environmental Microbiology, 5, pp. 1094-1102.
- Narasingarao, P., et al. (2012), "De Novo Metagenomic Assembly Reveals Abundant Novel Major Lineage of Archaea in Hypersaline Microbial Communities", The ISME Journal, 6, pp. 81-93.
- Nickle, D.C., et al. (2002), "Curiously Modern DNA for a '250 Million-Year-Old' Bacterium", Journal of Molecular Evolution, 54, pp. 134-137.
- Norton, C.F. and W.D. Grant (1988), "Survival of Halobacteria Within Fluid Inclusions in Salt Crystals", Journal of General Microbiology, 134, pp. 1365-1373.
- Oren, A. (2011), "Thermodynamic Limits to Microbial Life at High Salt Concentrations", Environmental Microbiology, 13, pp. 1908-1923.
- Park, J.S., et al. (2009), "Haloarchaeal Diversity in 23, 121 and 419 MYA Salts", Geobiology, 7, pp. 515-523.

- Pironon, J., et al. (1995), "Organic Inclusions in Salt. Part I: Solid and Liquid Organic Matter, Carbon Dioxide and Nitrogen Species in Fluid Inclusions from the Bresse Basin (France)", Organic Geochemistry, 23, pp. 391-402.
- Posey, H.H. and J.R. Kyle (1988), "Fluid-Rock Interactions in the Salt Dome Environment: An Introduction and Review", Chemical Geology, 74, pp. 1-24.
- Sass, A.M., et al. (2008), "Diversity of Bacillus-Like Organisms Isolated from Deep-Sea Hypersaline Anoxic Sediments", Saline Systems, 4, pp. 8.1-8.11.
- Sassen, R., et al. (1994), "Oligocene to Holocene Hydrocarbon Migration and Salt-Dome Carbonates, Northern Gulf of Mexico", Marine and Petroleum Geology, 11, pp. 55-65.
- Satterfield, C.L., et al. (2005), "New Evidence for 250 Ma Age of Halotolerant Bacterium from a Permian Salt Crystal", Geology, 33, pp. 265-268.
- Saunders, J.A. and R.C. Thomas (1996), "Origin of 'Exotic' Minerals in Mississippi Salt Dome Cap Rocks: Results of Reaction-Path Modeling", Applied Geochemistry, 11, pp. 667-676.
- Schléder, Z., S. Burliga and J.L. Urai (2007), "Dynamic and Static Recrystallization-Related Microstructures in Halite Samples from the Kłodawa Salt Wall (Central Poland) as Revealed by Gamma-Irradiation", Neues Jahrbuch für Mineralogie und Petrologie, 184, pp. 17-28.
- Schubert, B.A., et al. (2010), "Dunaliella Cells in Fluid Inclusions in Halite: Significance for Long-Term Survival of Prokaryotes", *Geomicrobiology Journal*, 27, pp. 61-75.
- Siemann, M.G. and B. Ellendorff (2001), "The Composition of Gases in Fluid Inclusions of Late Permian (Zechstein) Marine Evaporites in Northern Germany", Chemical Geology, 173, pp. 31-44.
- Sonnenfeld, P. (1995), "The Color of Rock Salt A Review", Sedimentary Geology, 94, pp. 267-276.
- Swanson, J.S., et al. (2012), Status Report on the Microbial Characterization of Halite and Groundwater Samples from the WIPP, LA-UR-12-22824, Los Alamos National Laboratory, Carlsbad. NM.
- Turkin, A.A., et al. (2006), "Radiolysis of NaCl at High and Low Temperatures: Development of Size Distribution of Bubbles and Colloids", Journal of Physics: Condensed Matter, 18, pp. 5655-5668.
- Urai, J.L. and J.N. Boland (1985), "Development of Microstructures and the Origin of Hematite in Naturally Deformed Carnallite", Neues Jarhbuch für Mineralogie Monatshefte, pp. 58-72.
- Vreeland, R.H., et al. (1998), "Distribution and Diversity of Halophilic Bacteria in a Subsurface Salt Formation", Extremophiles, 2, pp. 321-331.
- Vreeland, R.H., W.D. Rosenzweig and D.W. Powers (2000), "Isolation of a 250 Million-Year-Old Halotolerant Bacterium from a Primary Salt Crystal", Nature, 407, pp. 897-900.
- Wesełucha-Birczyńska, A., T. Toboła and L. Natkaniec-Nowak (2008), "Raman Microscopy of Inclusions in Blue Halites", Vibrational Spectroscopy, 48, pp. 302-307.
- Willerslev, E. and M.B. Hebsgaard (2005), "New Evidence for 250 Ma Age of Halotolerant Bacterium from a Permian Salt Crystal: Comment", Geology, 33, p. e93.
- Wolfram, J.H. and W.J. Dirk (1997), "Biofilm Development and the Survival of Microorganisms in Water Systems of Nuclear Reactors and Spent Fuel Pools", in J.H. Wolfram, R.D. Rogers and L.G. Gazsó (Eds.), Microbial Degradation Processes in Radioactive Waste Repository and in Nuclear Fuel Storage Areas, Kluwer Academic Publishers, Dordrecht, pp. 139-147.