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Identification and applicability of analogues for a safety case for a HLW repository in evaporites: results from a NEA workshop

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Abstract A workshop was held in September 2012 in Braunschweig, Germany, to discuss the potential for natural and anthropogenic analogue studies to contribute to safety cases for radioactive waste repositories constructed in salt formations. Presentations were given on many analogue sites and systems from different countries. Discussions at the workshop then addressed the following aspects that are particularly relevant to the safety concept for radioactive waste disposal in salt: (1) the long-term integrity of rock salt formations, (2) the integrity of technical barriers, and (3) microbial, chemical and transport processes. A diverse range of natural systems were discussed as potential analogues for the integrity of rock salt. These included the deformation of anhydrite layers in rock salt; the response of rock salt to mechanical and thermal loads; and the isotopic signatures of syngenetic waters contained in fluid inclusions. Some anthropogenic examples drawn from the oil and gas industries, and from hazardous waste disposal, were proposed as analogues for the integrity of (geo)technical barriers. A broad range of studies on natural and anthropogenic salt-brine systems were identified as potential analogues for the radionuclide sorption and (co)precipitation process that may take place in the repository near and far fields, as well as for understanding the significance of hydrocarbons and microbial processes. It was evident from discussions at the workshop that there are some specific technical issues that may benefit from further analogue study, particularly the compaction of crushed salt backfill, the viability of microbes in the near-field, the stability of plugs and seals, the deformation of anhydrite, and isotope signatures in fluid inclusions.

Keywords Nuclear waste disposal · Germany · Rock salt · Mechanical behaviour · Chemical processes · Microbial activity

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1 Introduction

In 2012, the Nuclear Energy Agency (NEA) of the Organization for Economic Co-operation and Development (OECD) in Paris established under the aegis of the Radioactive Waste Management Committee (RWMC), a new 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—to consider all aspects of radioactive waste (HLW and long-lived) disposal in rock salt. The Salt Club consists of representatives of the founding member states, namely Germany, USA, The Netherlands, and Poland. It 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 formation for a geological repository.

As one of its first activities, the joint workshop "Natural Analogues for safety cases of Repositories in Rock Salt" was held from 4th to 6th September 2012 in Braunschweig, Germany, hosted by the Project Management Agency Karlsruhe—Water Technology and Waste Management (PTKA-WTE) and the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH. The objective of the workshop was to compile studies about natural and anthropogenic analogues from different countries that may potentially be used within overall evaluations of long-term safety (i.e., "safety cases") for radioactive waste repositories in salt formations. A specific aim of the workshop was to identify and discuss studies that may provide underpinning and supporting knowledge of (1) the long-term integrity of rock salt formations, (2) the integrity of technical barriers, and (3) microbial, chemical and transport processes. The workshop was structured around the following topics:

- general aspects of analogues,
- status of disposal programmes in the Salt Club member states,
- integrity of rock salt,
- integrity of technical barriers,
- microbial, chemical and transport processes.

The workshop was attended by 37 scientists from eight countries, drawn from research institutes, universities, regulators and federal institutes as well as from companies actively involved in the salt mining and oil/gas storage industries. This paper summarizes the outcomes of the workshop, and illustrates and discusses selected contributions in some detail. More information about the workshop and each contribution can be found in Noseck and Miller (2013).

2 General aspects of analogue studies

There has been considerable development in thinking with regards to natural analogues over the last few decades. 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 rigor 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 "bottom-up" approach, Alexander et al. (2013) suggested that, in future, a 'top-down' method should be followed when identifying natural analogues. In this way, 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 is 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 modeling studies) to seek information to help confirm that a safety function is met. Many different natural systems and sites, from a number of countries, were discussed at the workshop. Several of these have not previously been recognized 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, the following 'types' of analogues may be identified in a safety case:

- 'industrial' analogues—such as gas storage caverns in rock salt, that may provide information on aspects to do with the behaviour of the rock during excavation of the repository and installation of the technical barriers;
- 'contemporary' analogues—that may provide information on short-term processes, such as the response of the rock mass during reestablishment of Thermal–Hydraulic–Mechanical–Chemical (THMC) equilibria or the investigation of healing processes after rock collapses in salt mines;
- 'operational' analogues—that provide information on practical aspects of constructing seals or performing backfilling excavations in salt, which are likely to be increasingly useful as disposal 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;
- 'self' analogues—that refer to information derived from an actual repository site, rather than from another location. This category of analogues overlaps with site characterization but their purpose is to provide information relevant to future system evolution rather than current site conditions.

It is not always useful to consider every type of analogue in a safety case. The choice and use of analogues should be considered in relation to the purpose of a safety case (e.g., concept development, design optimisation, formal licensing etc.) and the intended audiences for the safety case documents.

An important aspect that was stressed during the discussions at the workshop was the importance of correct and unambiguous terminology when discussing the possible 'types' of analogue studies. This is necessary to avoid possible misunderstandings between different audiences. This is an issue for the safety case as a whole because it should be recognized that the meaning of certain words used by the public in common speech is sometimes different to the usage by experts, and even occasionally between different groups of experts.

There can be differences of opinion in 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 to support development or qualification of safety assessment models, it should be of the highest quality. It is also vital 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 optimization work, and more quantitative information would be needed.

As suggested by Pescatore (2013), 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 identified the importance of terminology for confidence building.

3 Analogues for the long-term integrity of rock salt formations

The primary characteristics (and safety functions) of rock salt that contribute to long-term isolation of the waste from the accessible environment are very low hydraulic conductivity and the ability to deform plastically under stress, so that discontinuities that might present potential pathways can "self-seal". This is somewhat different to the characteristics of other potential host rocks that provide isolation primarily by processes such as radionuclide sorption, dilution and dispersion along groundwater flow paths. Therefore, an important contribution to fulfilling the safety function "containment of the waste" comes from the physical characteristics of the host salt rock formation itself. As a consequence, a fundamental part of the safety case is to demonstrate the integrity of the rock salt formation in the long-term. Here, analogues can play an important role. Relevant aspects that might impact the integrity are subrosion and diapirism; potential transport pathways in the geological barrier; mechanical load capacity (e.g., earthquakes and uplift with respect to competent rock formations); and thermal load capacity due to high temperatures produced by the waste or low temperatures as a consequence of glaciation (Wolf and Noseck 2013).

Mechanical processes in rock salt have been investigated at numerous sites and on different spatial scales. On a large scale, 3D seismic modelling of salt domes by the oil and gas industry described by Urai et al. (2013) showed the influence on the salt dome structure of the mechanically stronger anhydrite layers that are typically interbedded with halite. This is supported by laboratory scale studies such as those described by Mertineit et al. (2012), showing that, when subjected to stress, halite will deform by plastic flow whereas anhydrite will deform by brittle fracture. In a repository environment, such fractures may be possible pathways for groundwater flow, which is a key concern dealt with in the safety case. Detailed observations from boreholes at the Gorleben salt dome presented by Hammer et al. (2013) showed, however, that the anhydrite at Gorleben contains no large-scale interconnected porosity, but clear evidence of secondary mineralisation sealing the fractures. This is a good example of how laboratory, field and analogue studies can be combined to increase process understanding and, in particular, to address the issue of spatial scaling.

According to the German safety requirements (BMU 2010) it has to be shown that the formation of secondary water pathways, which might lead to in- or outflow of potentially contaminated brines within the "isolating rock zone" can be excluded. It also has to be shown that any pore water that may be present in the isolating rock zone does not participate in the hydrogeological cycle outside of that zone. Arguments to demonstrate that no fluids from adjacent formations have intruded into the Gorleben salt dome can also be derived from natural analogue studies using trace element profiles (e.g., Br and Rb) in the salt rocks, together with the chemical composition and isotope content of fluid inclusions (brines and gases).

The workshop contribution from Dulinski and Rozanski (2013) illustrated the potential of isotopes to characterize the origin of brines found in salt formations. It contained an overview of isotope investigations of water occurrences which have been performed over the past 40 years in four Polish salt mines (Wapno, Inowrocław and Kłodawa salt mines, located in north-central Poland, and Wieliczka salt mine located in southern Poland). These studies demonstrate that the salinity of mine waters is largely independent of water origin. Therefore, the chemical (elemental) composition of brines occurring in salt mines cannot solely be used as an indicator of the origin of water. Instead, environmental tracers (stable and radioactive isotopes) 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 the stable isotope composition of water (δ^2H and δ^{18}O) 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. An example is given in Fig. 1. Three main groups of water were identified in the Kłodawa mine on the basis of their isotopic composition (Dulinski and Rozanski 2013). The first group (green points) represents syngenetic waters. They are of Zechstein age and represents different stages of evolution of sea water during desiccation of shallow lagoons. The second group (red points) represent waters posing a potential risk for the operation of the mine. These are of meteoric origin and of different age. Most of them carry a significant evaporation signature but, with current knowledge, it is difficult to decide whether the evaporation took place before infiltration or after the water migrated into the mine openings. The third group (blue points) represents technological waters and waters of unknown origin. Technological waters are relatively easy to recognize. Usually they are formed by condensation of atmospheric water vapour or as drops on the connectors of pipelines. These waters may create characteristic erosion holes on the floor of mine corridors. Some waters belonging to this group are, however, difficult to explain. Their stable isotope composition may result from complicated, multi-stage isotopic exchange processes with atmospheric water vapour and/or crystallization water in salt minerals.

This study clearly demonstrated that the oxygen and hydrogen stable isotopes provide a possible way to determine the origin of waters in mines, and have a crucial role in the risk assessment associated with water inflows. With respect to analogues, this kind of analysis (together with other isotopic methods) is of value for investigating the long-term integrity of salt formations. Such methods can be used to show that salt (and other minerals and mineral layers in rock salt) remain unchanged since their deposition and so have been isolated from intrusion of waters from other sources. Evidence for this can come from minerals embedded in rock salt, in fluid inclusions in rock salt

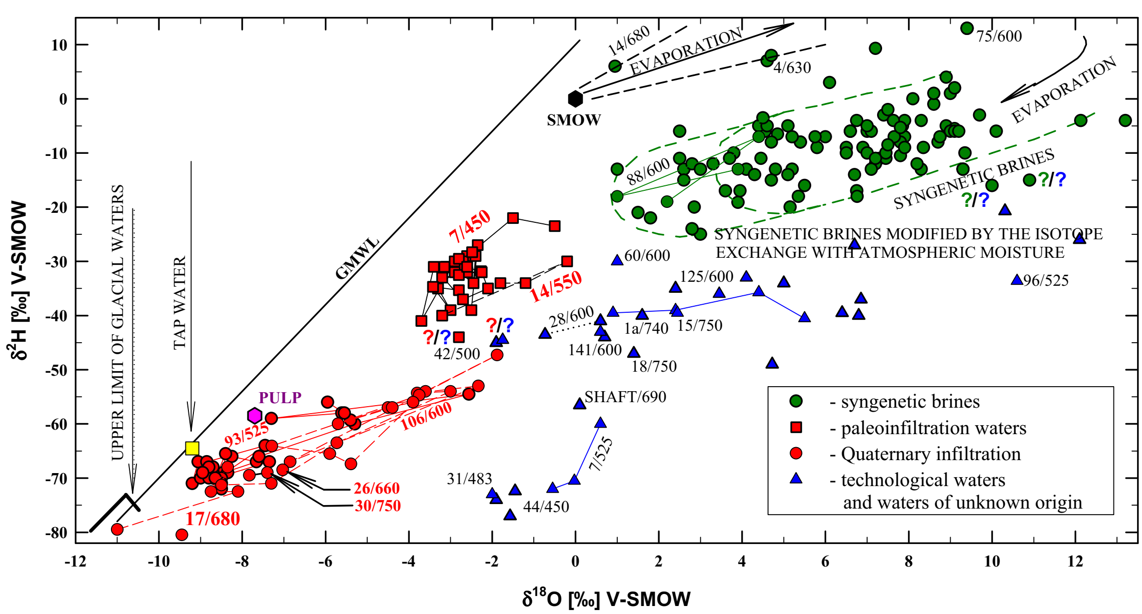

Fig. 1 Stable isotope composition of waters occurring in the Kłodawa Salt Mine (selected data, Dulinski and Rozanski 2013)

Table 1 Isotopic and thermometry methods used for long-term integrity analyses of salt domes

Method	Objective
$\delta^{18}O$ and $\delta^{2}H$ in brines of Zechstein rock salt	Distinction between formation waters of meteoric or marine origin
$\delta^{37}Cl$ in Zechstein rock salt	Extent of evaporation, input of seawater, re-dissolution of salt
$\delta^{34}S$ and $\delta^{18}O$ in sulfates of Zechstein rock salt	Indication of changes in the inflows, restriction conditions, redox reactions, and biogenic processes
$\delta^{13}C$ and $\delta^{18}O$ in carbonates	Post-sedimentation transformation of carbonates due to a contact with meteoric waters
$^{87}Sr/^{86}Sr$ in rock salt	Alteration of brine chemistry due to interaction processes with deep hydrothermal fluids or adjacent rocks
Micro-thermometry of fluid inclusions	Homogenization temperature: temperature during formation of fluid inclusion

minerals, and in brines from salt mines. Table 1 lists the various isotopic methods that have been used and their objectives.

4 Analogues for the long-term behaviour of technical barriers

Although the geotechnical barriers play a key role in achieving the safety function "containment of the waste" only a few workshop contributions dealt with this type of analogue application. The workshop therefore identified this topic as one where further analogue studies way be warranted.

An understanding of the nature of the boundary conditions and thresholds at which rock salt and its technical barriers might become negatively impacted comes from experience from other large-scale excavations in salt, such as mines and gas storage caverns, as described by Minkley and Knauth (2013) and by presenters from industry. 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 in (or influences on) boundary conditions have caused a loss of integrity, such as collapse of excavations, damage to surface structures, and groundwater flow into tunnels. Modeling studies show that these situations may arise if either the minimum stress or dilatancy criteria are exceeded.

Experiences from the disposal of hazardous wastes in salt and potash mines were described in Noseck et al. (2013). These industrial analogues provide 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 emplaced into the salt). These analogue sites may provide unique opportunities to help support the design of seals and plugs for a radioactive waste repository, and potentially also for performance confirmation monitoring of the seals over several decades into the future. Although clearly very informative, the transferability of industrial analogue information to repository systems needs to be carefully assessed. As is the case with all forms of analogues, the similarities and the differences between the conditions at an industrial analogue and those anticipated at a repository need to be identified and evaluated.

For example, there may be different views regarding the stability of rock salt, and its integrity as a barrier, due to the very different timescales over which it needs to be assessed for radwaste disposal compared to other industrial situations (e.g., mining). As a consequence, it cannot be assumed that practices (e.g., modeling approaches and facility designs) adopted in common industrial situations can be directly applied to the repository. They can, however, provide a very useful starting point, and a way to avoid duplicating effort where there are issues of common interest to both radioactive waste disposal and other industries that work in rock salt. As a consequence there should be clear benefits to be gained from cross-industry liaison, shared research, and exchange of knowledge and experience, to improve conceptual and numerical models for rock salt behaviour and to assess its integrity as a barrier over the long-term.

5 Analogues for microbial, chemical and transport processes

The workshop contributions concerning microbial, chemical and transport processes are mainly relevant to radionuclide release scenarios. These contributions describe studies on various topics including radionuclide retardation and precipitation, the importance of hydrocarbons in salt rock, and the occurrence and role of microbes in saliferous systems. Two presentations are discussed here that illustrate the possible application of analogues in this area.

Releases of Ra-226 often contribute to the calculated post-closure doses in safety assessments—particularly for repositories containing spent fuel—due to an assumption of high mobility, and the corresponding choice of conservative values for solubility and retardation factors in transport models. As explained by Metz et al. (2013), however, laboratory studies performed during recent decades have shown that Ra can often be retarded by co-precipitation with Ca and Ba within $(Ba,Ra)SO_4$ (barite) solid solutions

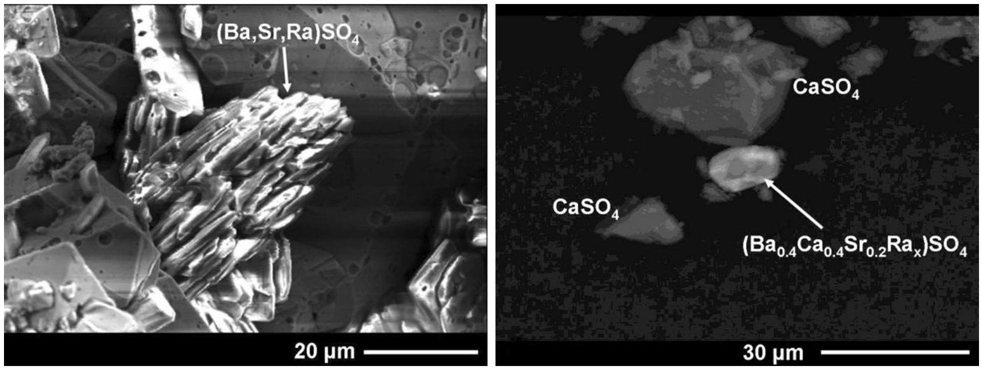

Fig. 2 SEM images of ternary and quaternary Ra-bearing barite solid solutions (Metz et al. 2013). *Left* image (Ba,Sr,Ra)SO_4(s) crystal in a precipitate sampled from a geothermal energy plant in Southern Germany; *right* image (Ba,Ca,Sr,Ra)SO_4(s) crystal precipitated in pond #1 of the K'tziot desalination facility, Southern Israel (Metz et al. 2013)

in both dilute and highly saline systems. These laboratory scale experiments can be underpinned by observations from man-made analogues such as large-scale evaporitic systems to support the application of laboratory results on formation of (Ra,Ba)SO_4(s) to the radium retention processes in repository systems.

This type of investigation is of interest, because it is expected that the formation of (Ra,Ba)SO_4(s) solid solutions (Fig. 2) reduces the maximal aqueous Ra concentration by several orders of magnitude in comparison to the solubility with respect to a pure $RaSO_4$(s) end-member (Grandia et al. 2008; Prieto et al. 2013). The investigations at the evaporation system in K'tziot desalination plant (near Nitzana in the Israeli Negev desert), shown by Metz et al. (2013), demonstrate that Ra was effectively removed through the precipitation of a (Ra,Ba)SO_4(s) solid solution; either as a binary solid solution, or as a part of a ternary (Ra,Ba,Sr)SO_4(s) solid solution showing compositional zoning with zones that are Ba-rich and zones that are Sr-rich. The results of this field scale evaporation study were comparable to those of the laboratory based experiments, suggesting that both in the complex field/natural system, and in the laboratory, the same factors affect the formation of the radiobarite solid solution. However, differences in partition coefficients measured in co-precipitation and re-precipitation experiments, respectively, need to be resolved in further studies, particularly to establish the extent to which the radiobarite co- and re-precipitation systems approach near-equilibrium conditions (Metz et al. 2013).

The potential for using natural analogues to assess whether microbial activity might impact the properties and/or processes in a repository system in rock salt, and the consequences for post-closure safety, was reviewed by Meleshyn (2013) who gave an overview of experimental findings and field studies.

Meleshyn showed examples from different studies in salt formations, where the existence of microbes was claimed (see Fig. 3). There is no doubt that ancient halite deposits support substantial populations of halophilic bacteria and haloarchaea (Grant 2004). Such microbes might have survived for extremely long time scales. For example, the 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) has been reported (Vreeland et al. 2000). Metabolic activity—even at very low levels such as for DNA repair—requires an in situ energy source. This poses a further question on the mechanism of microbial survival in fluid inclusions. Both the potential sources of energy and the survival of microbes in fluid inclusions were discussed by Vreeland et al. (2000). An example from Death Valley showed that microbes in fluid inclusions with an age of 12,000–100,000 years were often observed to be co-trapped with the *Dunaliella* algae, which contain up to 7 M glycerol, $C_3H_5(OH)_3$, in their cytoplasm and release this source of energy into brine (Schubert et al. 2010). Although virtually all hypersaline lakes contain large

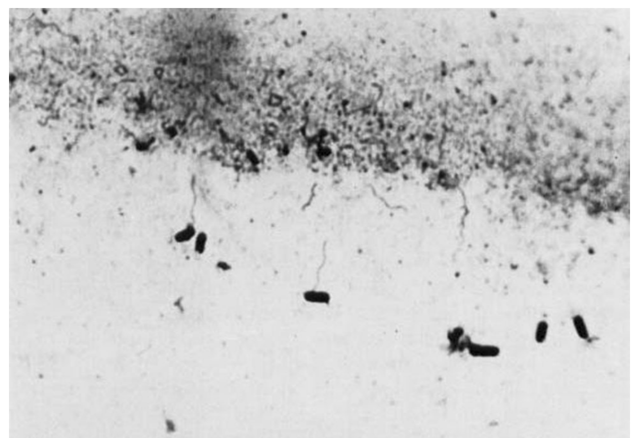

Fig. 3 Microbes in the Precambrian salt (Meleshyn 2013: source Dombrowski 1963)

amounts of dead plant material, the Salado formation contains no sign of fossilized organics but detectable populations of cellulose-degrading microbes (Vreeland et al. 1998). This suggests that microbes may survive in rock salt over long periods of time by utilizing co-deposited organics as energy and nutrient sources.

Therefore, it is probable 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 so 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. Even if laboratory and field studies (e.g., at WIPP) confirm the presence of ancient indigenous microbes as well as recent introduced species, few analogue studies have provided convincing evidence for their viability under repository post-closure conditions in rock salt, e.g., in saturated brines at high temperatures.

6 Conclusions

The workshop illustrated the potential for analogue studies to be used in a safety case for a deep geological repository in rock salt. Most contributions focused on the integrity of the salt formation, which represents the main barrier for the containment of the waste. These studies addressed mechanical properties (e.g., the role of mechanically stronger anhydrite layers in rock salt as potential fluid pathways), the behaviour of rock salt against mechanical and thermal loads, and isotopes in fluid inclusions as indicators for the origin of water in salt formations. To date, few analogue studies of the integrity and long-term stability of (geo)technical barriers have been identified. Studies from the oil and gas industries, and from hazardous waste disposal, have the potential to contribute as analogues for such barriers. Importantly, the workshop identified analogue studies for radionuclide retention and precipitation processes, as well for microbes in rock salt, which are mainly relevant for release scenarios. In the final discussion of the workshop it was stated that, with respect to the safety case, the topics that may benefit most from further analogue investigations are (1) the compaction of crushed salt as a technical barrier, (2) microbial activity in salt, (3) deformation of anhydrite, (4) fluid inclusions, and (5) gas storage. An open discussion involving the radwaste community and scientists from other disciplines and industry was recommended.

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