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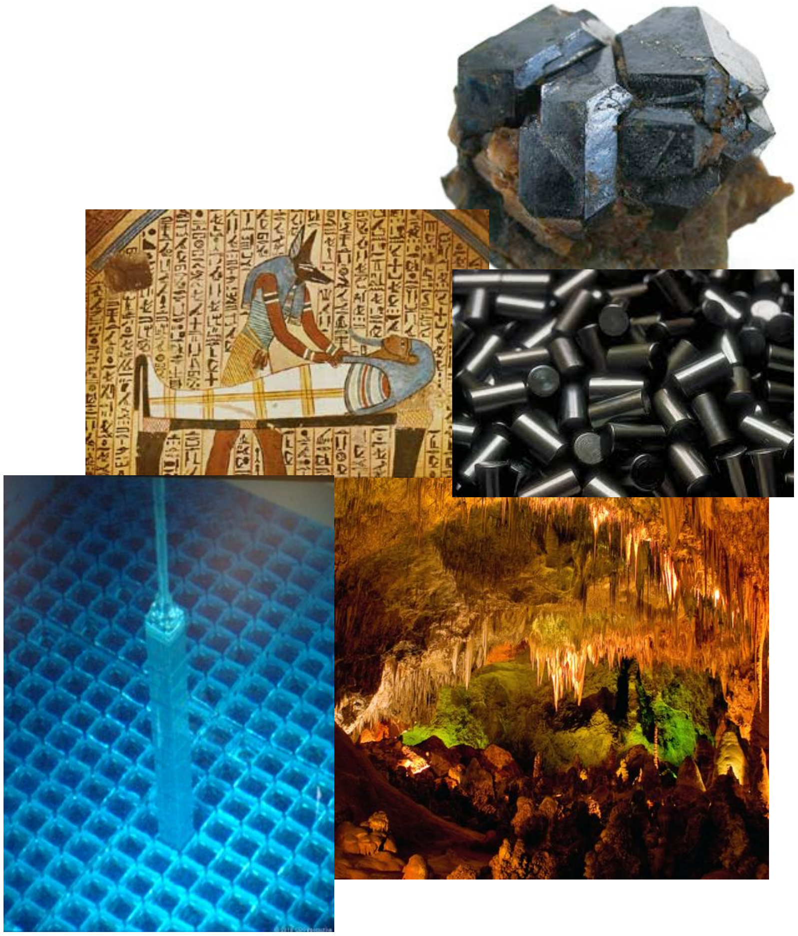
Natural and Anthropogenic Analogues for High-Level Nuclear Waste Disposal Repositories: A Review

A report to the Canadian Nuclear Safety Commission (CNSC)



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Natural and Anthropogenic Analogues for High-Level Nuclear Waste Disposal Repositories: A Review

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Acronyms

CNSC	Canadian Nuclear Safety Commission
DGR	Deep Geological Repository
DoE	Department of Energy
EBS	Engineered Barrier Systems
EC	European Commission
HLNW	High Level Nuclear Waste
IAEA	International Atomic Energy Agency
IRB	Iron Reducing Bacteria
NAWG	Natural Analogue Working Group
NB	Natural Barriers
NRC	Nuclear Research Council
OCRWM	Office of Civilian Radioactive Waste Management
REE(s)	Rare Earth Element(s)
SA	Safety Assessment
SRB	Sulfur Reducing Bacteria
UNF	Used Nuclear Fuel
USGS	United States Geological Survey

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ABSTRACT

This report provides a review of natural and anthropogenic analogues for high-level nuclear waste disposal in a deep geological repository (DGR). We also occasionally highlight analogues that have been used for low and intermediate waste. Most studies define natural analogues as either naturally occurring or anthropogenic (manmade) systems. In this report, we distinguish between natural analogues and anthropogenic analogues because anthropogenic analogues generally provide non-technical (anecdotal) illustration of concepts and processes for the safety case, whereas natural analogues can provide technical and quantitative information. In addition, natural analogues can provide information over geological time- (millions of years) and spatial- (kilometers) scales whereas anthropogenic analogues provide information over a much more limited time-scale (hundreds or thousands of years). Regardless of the definition, analogue studies provide one of the multiple lines of evidence intended to increase confidence in the safe geologic disposal of high-level nuclear waste. They are deemed necessary because they complement the experiments that are carried out over a period of months or years. They also provide a way to validate numerical long term safety assessment models with information and data covering geological time- and spatial-scales.

The first part of this report describes the analogue concept. The second and third parts provide examples of natural and anthropogenic analogues for engineered barrier systems (EBS) and natural barriers (NB). Part four describes analogues for complex coupled transport processes and finally we provide general and specific recommendations for future research in part five of the report.

A key recommendation is that a concerted effort should be made to ensure that there is a transfer of data from the complex, natural analogue field studies to simplistic models which, by necessity, are used in safety assessments (SA). Field analogue studies should be planned to interface with laboratory experiments and, ultimately, with *in situ* field experiments, when the final repository site is selected. This will involve using natural analogue data in a quantitative way to support the DGR safety case.

1. INTRODUCTION

With increasing concern over climate change due to the consumption of fossil fuels, identifying clean sources of energy are of paramount importance. Nuclear energy is one of the cleanest sources of energy. Although countries such as France depend on nuclear power for ~80% of their energy needs, other countries have been slow to embrace this source of energy. In Canada, 15% of its electrical energy needs comes from nuclear power where most of the capacity is in Ontario, which derives 50% of the provincial energy needs from nuclear power. One the major stumbling blocks in the wide-spread use of nuclear energy is related to the safe disposal of long-lived and highly-radioactive nuclear waste (i.e. used nuclear fuel) generated by this form of energy. Although several options have been proposed for the disposal of radioactive waste, there is a global consensus that the safest method for the disposal of high-level nuclear waste (HLNW), which in most cases largely consists of solid, used nuclear fuel (UNF), is in a subsurface deep geological repository (DGR; Miller, 2000). A DGR utilizes a multi-barrier approach (engineered and natural) to restrict the rate of release of radionuclides over extended periods of time (Fig. 1).

Given enough time, natural radioactive decay of HLNW will reduce the radioactivity to levels similar to naturally occurring uranium mineral deposits. One of the challenges facing the nuclear industry is to demonstrate that a subsurface DGR will contain and isolate the waste for a sufficient amount of time allowing natural radioactive decay to render the HLNW relatively safe. It is generally accepted that after 1 million years (e.g., IAEA 2012) radioactive decay of nuclides (e.g., neptunium, Np) in HLNW will reduce their inventory to sufficiently low-levels that any radionuclide release will pose no significant health or environmental risk.

Natural barriers (i.e., the geosphere) can isolate HLNW for extended periods of time (e.g., millions of years) from the biosphere, and limit the impact of external perturbations such as groundwater incursion, and limit access to fast radionuclide transport pathways (e.g., faults and fractures). The geosphere can also reduce the rate of degradation for engineered barriers and HLNW by providing a stable physical and chemical environment within the repository, attenuating radionuclide transport by virtue of its hydraulic and sorptive properties (CNSC, 2006).

In Canada, the safety of a DGR is assessed using a concept called the safety case. A DGR safety case is built along multiple lines of reasoning, consisting of geological information, information about the engineered barrier system, container corrosion, the waste-form characteristics, and the safety assessment models (CNSC, 2006). Any project undergoing an environmental assessment will consist of safety assessments to make predictions about a project's impact over its operation. However the safety case concept applies to repositories, because of the very long time frames involved. A long term safety assessment often considers a scenario of expected evolution of a site or facility to predict: (1) contaminant release, (2) contaminant transport, (3) receptor exposure, and (4) potential effects resulting from the exposure. The safety case for long-term disposal of HLNW in a DGR relies on experimental and geological studies to demonstrate system robustness.

Geological studies quantify several processes that may affect the repository such as groundwater flow rates and consequent radionuclide transport. These studies include geophysical, geochemical, and hydrogeological information, including paleohydrogeology. Geological information is unique in that it can offer lines of reasoning that span millennia. It can also offer multiple and independent lines of evidence (qualitative and quantitative) to support a

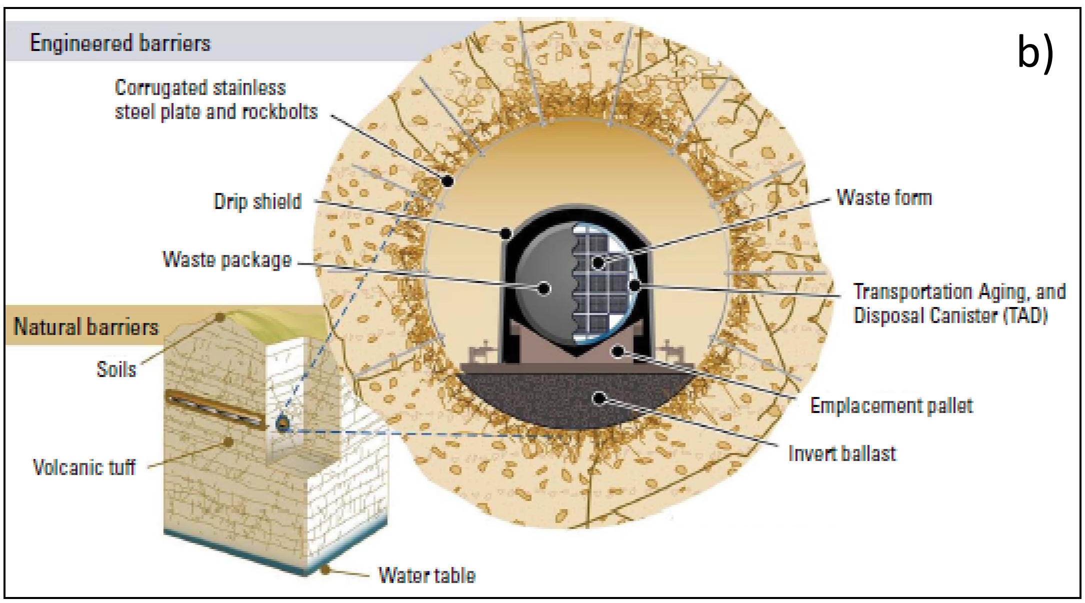


Figure 1. a) Photograph of the Yucca Mountain Project (YMP) site and b) image showing the engineered and natural barriers associated with the YMP DGR concept. Images are from Wikipedia and USGS (2010), respectively.

safety case. Furthermore, geological information can be used to select the appropriate site and design for a repository (CNSC, 2006).

One of the more difficult concepts involved in the disposal of radioactive waste in DGRs are the very long time frames required by the safety assessment. This is well beyond anything that can be considered in an experimental setting (e.g. large-scale projects in underground laboratories and laboratory experimental mockups). Mathematical models that make long term estimates face considerable uncertainties over these time frames. Due to the uncertainty of predictions made far into the future (e.g., 1 million years) the reliability of quantitative predictions (experimental data) diminishes with increasing timescale. Therefore, the demonstration of safety will rely less on quantitative predictions and more on qualitative arguments as timescales increase. Natural analogues can complement numerical modeling with quantitative data from systems that formed over geologically long time periods, and that have remained stable for millions to billions of years.

System robustness can be demonstrated using natural analogues. Most studies define natural analogues as either naturally occurring or anthropogenic (manmade) systems in which processes similar to those expected to occur in a DGR are thought to have occurred over long time periods (decades to millennia) and large spatial scales (crustal-scale) (Petit, 1990). Analogues used in the evaluation of a potential DGR provide an important temporal and spatial dimension to the understanding of processes and events that may take place in or around a repository (USGS, 2010).

Natural analogue studies have been widely used internationally by organizations such as the European Commission’s (EC) and Natural Analogue Working Group (NAWG; IAEA, 1989, 1999; Miller et al., 2006) to build confidence in the ability of waste management systems to perform over the long term as predicted by safety assessment models. For example, the International Atomic Energy Agency (IAEA) stated that:

“The natural analogue is often regarded as one of the very few means by which it may be possible to demonstrate to the public that safety assessments are based on a realistic understanding of how nature works over time periods longer than the existence of mankind.” (IAEA, 1989). The IAEA and the EC’s NAWG noted:

“The role of a natural analogue should be to confirm: (a) that the process is, in fact, something which can or will occur in practice, as well as in theory, and in nature, as well as in the laboratory; (b) where, when, and under what conditions it (the process) can occur; (c) that the effects of the process are those envisaged in the model; and (d) that the magnitude of the effects, in terms of scale and time, are similar to those predicted for a similar set of conditions” (Chapman and Smellie, 1986).

More recently, the use of analogues in communicating with the public has been viewed as an important part of the process for building confidence in the DGR concept (Stuckless, 2002, 2003, 2006; Lopez et al., 2004). Therefore, natural analogues can be used to demonstrate that waste management concepts actually work in nature. Natural analogues can also be the subject of complementary assessments of long-term safety, and can be included in the safety case to provide confidence in safety assessment models. Information from natural analogue studies can be used to develop and test detailed processes that may be incorporated numerical models in a simplified manner. Natural analogues can also provide data for developing generic site descriptive models in the absence of site-specific characterization data.

This report summarizes our current state of knowledge regarding analogue systems that can be used to demonstrate DGR system robustness. The report is divided into five sections. Sections 2 and 3 discuss analogues for engineered and natural barriers, respectively. Section 4 describes analogues for coupled transport processes and section 5 provides some recommendations for future work.

1.1. Analogue Concepts

As stated previously a DGR is complex facility, which consists of natural and engineered barriers (Fig. 1). An analogue study can investigate any part or process relevant to the DGR, to provide qualitative or quantitative information that can support and build confidence in a geological repository (Miller, 2000). Analogue studies can, therefore, provide data that is directly applicable to a safety assessment model (e.g., parameter values) or may provide illustrations of concepts or processes as lines of evidence to be included in the safety case (Miller et al., 2006). The latter is generally non-technical and useful for communicating concepts and processes to the public.

In much of the literature (Miller, 2000; USGS, 2010), natural analogues are conflated with anthropogenic analogue studies. Studies generally focus on a small number of processes (Miller, 2000; USGS, 2010). Miller (2000) defines analogue studies into three broad groups: (1) natural geological and geochemical systems; (2) archaeological systems; and (3) sites of anthropogenic contamination. In our report, we distinguish between natural and anthropogenic analogue studies because they provide information on different time and spatial scales (Fig. 2) and can be used to model different types of barriers within a repository. For example, anthropogenic studies can be used to study degradation rates of engineered barriers (e.g., cementitious material) whereas natural analogues (e.g., uranium deposits) can be used to study corrosion of the source-term and radionuclide transport rates over geological time scales.

1.1.1. Anthropogenic Analogues

Anthropogenic analogues also referred to as *archaeological* or *industrial* analogues provide non-technical illustrations of concepts and processes for the safety case (Miller and Chapman, 1995; Miller, 1996, 2000). Anthropogenic analogues are particularly useful for providing insight into the decay of engineered barriers. For example, progressive decay of human-made artifacts (e.g., tools, jewelry, copper cannons) can provide information on the "long-term" behavior of repository material such as cementitious build-out material, steal infrastructure, and canisters and casings. Although the time period of study is constrained to a few hundred or thousand years (Fig. 2), these human-made systems can provide valuable information on the rates of processes relevant to the early-life of a repository (Miller, 2000).

A particular advantage of analogues studies is they can provide non-technical and visual illustration of the basic concepts of repository design and performance for non-technical (public) audiences. Common problems with using anthropogenic analogues are the environments that preserve the artifacts are often not similar to subsurface repository conditions, and there is a sampling bias. For example, museums often house the best preserved artifacts. Therefore, corrosion rates based solely on museum artifacts may not be conservative (Miller, 2000).

1.1.2. Natural Analogues

There are several natural geological and geochemical systems that can be studied as analogues for different systems within a DGR. The most common natural analogues are: (1) uranium deposits and associated clay minerals; (2) naturally occurring cements and high-pH fluid systems; and (3) naturally occurring metals, glasses and bitumens. Fluid-rock interaction is

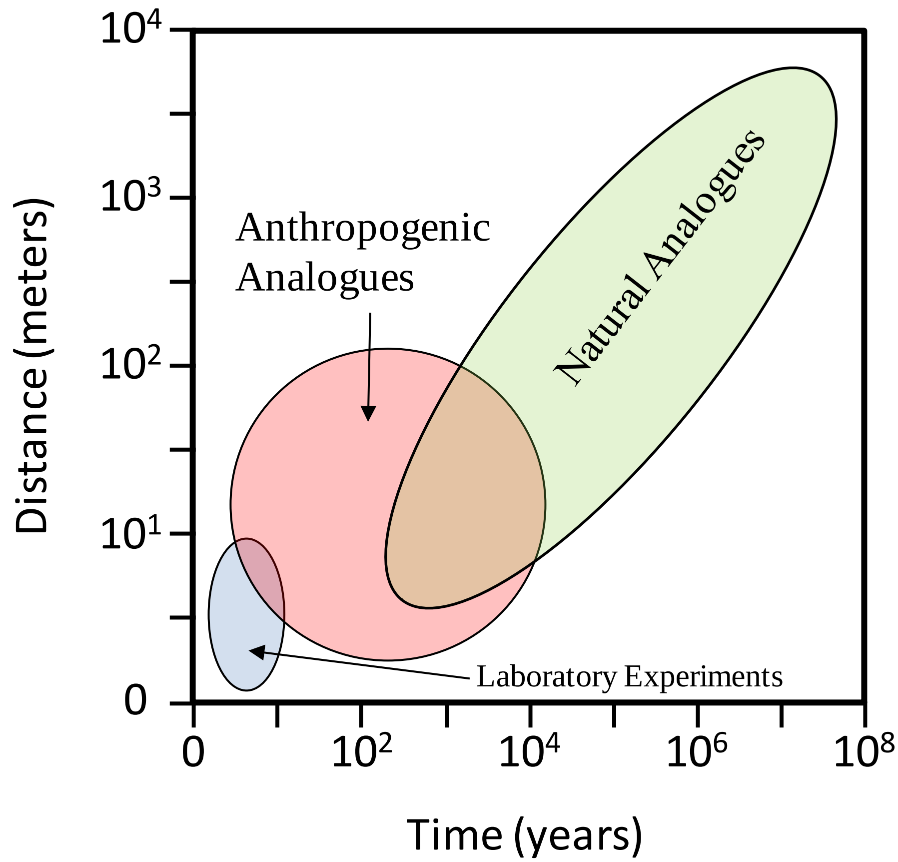


Figure 2. A comparison of time and spatial scales for laboratory experiments (days to months), anthropogenic analogues (hundreds to thousands of years) and natural analogues (e.g., >1 million years).

a common underlying theme for all three natural analogues. Therefore, it is necessary to study the hydrothermal systems associated with each analogue, which will provide information on the physical and chemical properties (e.g., minerals in fractures and pore-spaces) of near-field rocks, which may affect radionuclide transport and sorption, solubility and speciation of radionuclides, and chemical and isotopic gradients associated with veins (Miller, 2000; USGS, 2010).

Uranium deposits can be used as analogues for several systems within a DGR. For example, uranium ore (Fig. 3) is an excellent analogue for the UNF source-term and near-field processes associated with HLNW (e.g., Janeczek et al., 1996; Fayek et al., 1997a,b). Secondary deposits and remobilized ore (Fig. 4) are of great interest because they usually form from low-temperature fluids (<100^{o}C), which are representative of repository conditions (USGS, 2010). Therefore, uranium deposits can provide information on: (1) the corrosion or dissolution rates of uraninite ($UO_{2.x}$; a common uranium ore mineral with similar chemistry and physical properties to UNF) as an analogue for UNF; (2) reduction-oxidation (redox) processes in mobilizing and sequestering radionuclides; (3) speciation and solubility controls of radionuclides in groundwaters, including colloidal transport; (4) sorption, diffusion and rock matrix porosity; and (5) geochronology to determine the rates of mineral dissolution and element remobilization along fractures and faults (Miller, 2000; Fayek et al., 2006; Bruno and Ewing, 2006; USGS, 2010). Uranium deposits are particularly valuable because they can provide analogue information over geological time-scales (>1 billion years) and crustal length-scales (>500 meters).

The clay minerals that are often associated with uranium ore systems are analogous to the clay backfill or buffer in a repository, and the faults and fractures can provide important information regarding fast-pathways for radionuclide transport to receiving environments over geological time-scales and crustal length-scales. Clay minerals associated with uranium ore systems often occur along factures and faults, and are an alteration mineral that formed when a hydrothermal fluid interacted with the host-rock. Therefore, clay minerals can provide valuable information on: (1) diffusion and advection and small-scale heterogeneities; (2) element diffusion coefficients; (3) redox conditions and the pH of the fluid; (4) thermal stability and effects related to radiation; and (5) chemical complexing behavior of radionuclides and other elements in pore spaces and fractures (e.g., Kotzer and Kyser, 1995; Miller, 2000).

Cementitous material would be present in repositories in the waste packaging, construction material and backfill. In Jordan, natural cements formed from high temperature metamorphism of bituminous marls and limestone, caused by spontaneous combustion. Fluid interaction with cementitious zones would generate high pH fluids that could react with host rocks and barrier material to produce variable and potentially predictable fluid and mineral alteration compositions and sequences (e.g. Savage, 2011). Natural fluid systems with high alkalinities may be analogous to porewaters in cements and concretes that may be used as build-out material in a DGR. These fluid systems are relatively rare in nature and often involve water/rock interaction involving an ultramafic system (e.g., Oman analogue site). The study of these alkaline systems provides information on the solubility and speciation of radionuclides at high-pH conditions, interaction of high-pH fluids with surrounding host-rock, which may be analogues to porewater migration from the repository into the host-rock, and microbial mediated processes at high-pH, such as waste-form breakdown and near-field radionuclide mobilization (Miller, 2000).

Naturally occurring repository materials are generally found as isolated and often uncommon occurrences in natural. These include natural volcanic glass, native metals (e.g., copper), bitumen and natural cements. The study of naturally occurring volcanic glass and native

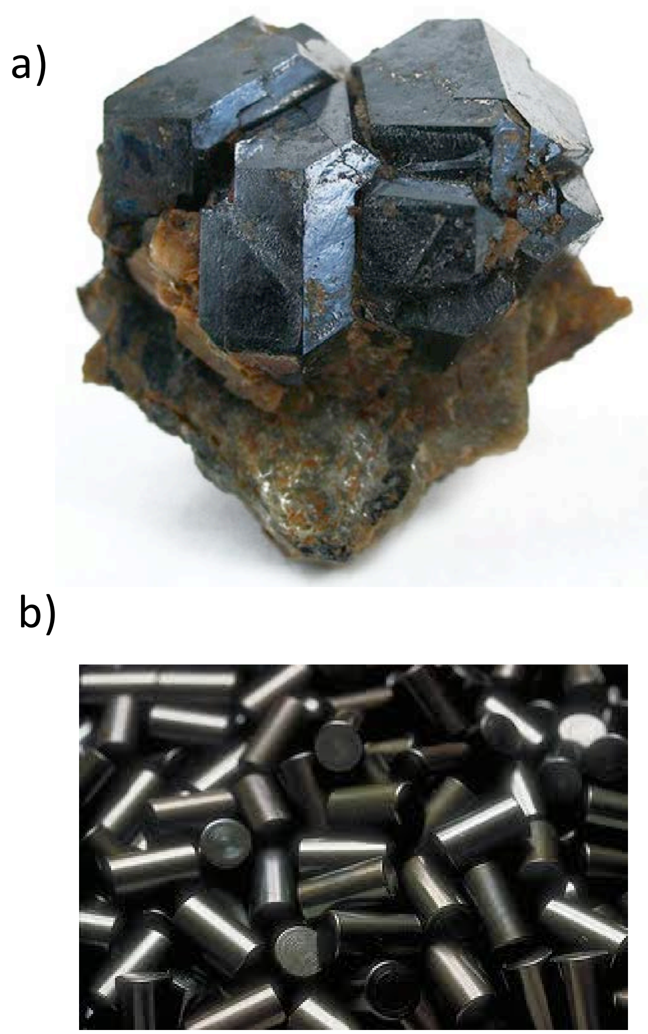


Figure 3. a) Uraninite crystals from the Topsham mine, Maine, USA, and b) UO_2 fuel pellets. Images are from Wikipedia and Scholle and Ulmer-Scholle (1997), respectively.

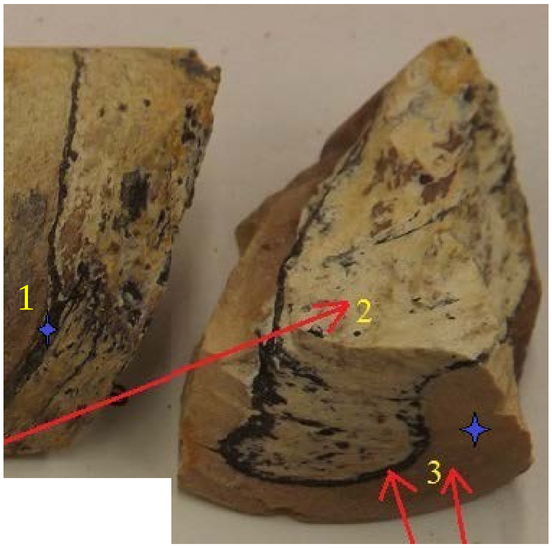


Figure 4. Image of a core samples from the End uranium deposit, Kiggavik area, Nunavut, Canada. Area 1 shows uranium minerals along a fracture, and areas 2 and 3 show secondary uranium mineralization within a clay rich matrix.

metal corrosion can provide information the corrosion rates for glass wasteforms and canisters, respectively. The decomposition of naturally occurring bitumen is analogous to the degradation of bitumen associated with low-level and intermediate-level waste. The study of natural cements can provide valuable information (e.g., composition) related to the stability of proposed build-out material (Miller, 2000).

2. ANALOGUES FOR ENGINEERED BARRIERS AND REPOSITORY INFRASTRUCTURE

Uranium deposits are generally considered good natural analogues for several aspects of a DGR. These include, source term (UNF) dissolution rates, secondary mineral formations, clay back fill behavior and its effects on radionuclide immobilization, and hydrogeological and tracer studies. Anthropogenic analogues are often used to study waste package and build-out material degradation (e.g., canisters and cements), and repository infrastructure stability (e.g., underground opening). However, there are geological systems that are analogous to cementitious and bentonitic barrier systems (notably, the Maqarin analogue in Jordan). The following sections will summarize analogues that have been used to build confidence in the DGR concept.

Most SA models of the long term evolution of the multiple barrier system are conservative, considering potential exposure pathways that might occur in different scenarios. “Normal” evolution scenarios assume some degradation of the engineered barrier system (waste container + sealing material such as cement and bentonite) based on the characteristics of the waste, the container design, the sealing material and the host rock. In addition to the normal evolution, the evolution of a DGR system under disruptive scenarios are also considered, to include an assessment of the repository system in cases of events that are thought to be possible yet unlikely to occur, such as the presence of an undetected waster-conducting vertical fault close to the repository. Natural analogues can be used to evaluate and demonstrate the outcome of normal and disruptive events. For example, the waste package will degrade, and some containers will be breached at some point during the lifetime of the repository, releasing radionuclides via air and fluids upon interaction with the waste form (Fig. 5). This will occur because most of the many waste-forms are thermodynamically unstable under wet, oxidizing conditions. For example, used nuclear fuel (UO_2) will oxidize and hydrate, and glass waste forms will form clays, zeolites, and oxides (USGS, 2010). The rate of radionuclide release will depend on many factors, such as when degradation occurs during the lifetime of the repository. If the failure occurs early in the lifetime of the proposed repository, the waste-form will be highly radioactive and this radioactivity will generate heat that could create a hydrothermal cell. At this stage water may enter the waste package as a liquid but more likely interact with UNF in vapor form (USGS, 2010). Radiolysis (radiation-induced decomposition) of the humid air within the package may cause production of nitric acid, which could condense into liquid water. If the breach is late in the repository history, radiation levels and heat will be much lower, and evaporation and radiolytic acid production will be less of a factor (USGS, 2010). Therefore, a breach that occurs late in the repository history will release fewer radionuclides due to reduction of inventory from radioactive decay and cooler temperatures, which reduces the reaction rates. However, lower temperatures may increase the chance of microbially-induced degradation of the waste-form (Lovley et al., 1991). Consequently, it is very important to build a DGR in an environment and using materials that will minimize groundwater ingress, reducing the risk of an early breach of the waste-package.

Isolation and containment of radioactive waste within a DGR would be accomplished through a combination of natural features and engineered barriers. This complex system is

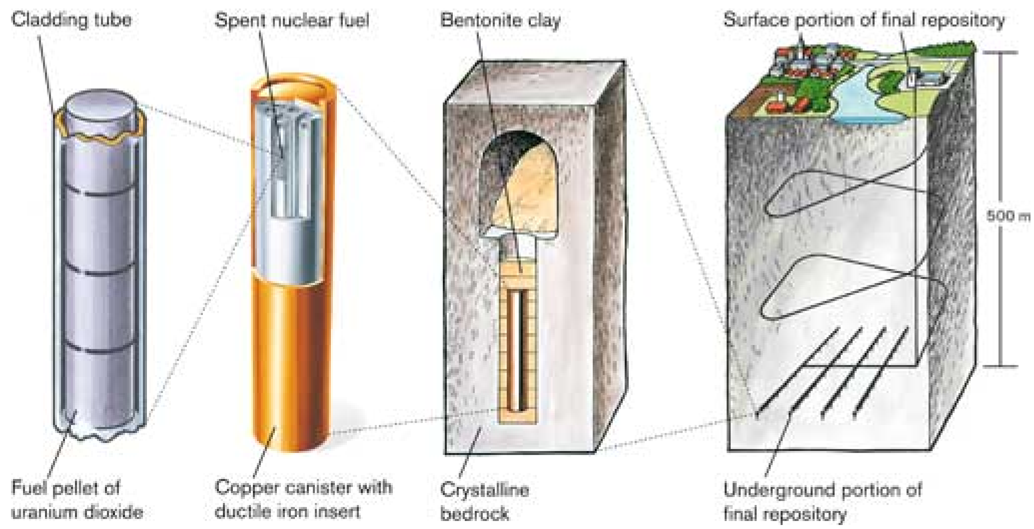


Figure 5. Cartoon illustration of used nuclear fuel pellets within a fuel assembly, waste package and DGR environment. Image is from IAEA (2012).

difficult to model and extrapolate data, especially from short-term experiments to times-scales of 1 million years or more. For this reason, the international community initiated several multinational analogue projects. Most analogue projects have focused on UNF as a source term because this is deemed to represent the highest risk to human health and the environment, and will constitute the majority (~90%) of the waste in a repository (USGS, 2010).

Several important and successful natural analogue programs have been sponsored by government agencies in both Europe (European Commission's (EC) natural analogue working group (NAWG)) and the United States (Department of Energy (DoE) and Nuclear Research Council (NRC)). The DOE Office of Civilian Radioactive Waste Management (OCRWM) has funded several natural analogue studies at Oklo, Gabon; Poços de Caldas, Brazil; Alligator Rivers, Australia; and Cigar Lake, Canada (Laverov et al., 2008; USGS, 2010).

One of the major findings from these studies is that analogues must be used with care so as to represent correctly the process or feature of interest (USGS, 2010). For example, in oxidizing conditions uranium occurs in the hexavalent state, which readily forms complexes with a number of ligands that are soluble and therefore mobile, whereas under reducing conditions (low dissolved oxygen) uranium occurs in a tetravalent state and forms oxides, which precipitate in the form of oxide minerals ($UO_{2.x}$; Romberger, 1984). Therefore, knowing the behavior of used fuel and glass waste forms under relevant repository conditions is essential because the dissolution and alteration of uraninite under reducing conditions is different from that under oxidizing conditions (Romberger, 1984; Janeczek and Ewing, 1992).

2.1. High-Level Nuclear Waste (Source Term)

Several analogues have been used to study: (1) used fuel dissolution rates, (2) immobilization of radionuclides by secondary minerals, (3) radiolysis, (4) criticality, and (5) glass waste forms. Uranium deposits can provide important information on the performance of radioactive waste forms and radioactive waste repositories (Janeczek et al., 1996) because uraninite (UO_{2+X}), the most abundant uranium-bearing mineral in most uranium deposits, is similar in many ways to the UO_2 in used nuclear fuel (Janeczek et al., 1996; Ewing, 1999). The uraninite structure can accommodate some degree of oxidation in highly oxidizing aqueous environments (Romberger, 1984). However, in highly oxidized environments, uraninite is unstable and decomposes (Finch and Ewing, 1992a). Secondary uranyl (U^{6+})-bearing phases form on the surface of the uraninite, and a rind of corrosion products forms (Fig. 6). A number of studies have examined the effects of impurities in uraninite on the rate of uraninite alteration and the composition of the corrosion products (Finch and Ewing, 1991; Fayek et al., 1997a,b; Fayek et al., 2000). Studies that have examined the dissolution of uraninite under oxidizing conditions have concluded that the dissolution rate were lower for uraninites that have higher concentrations of thorium, lead, and rare earth elements (REE; Finch and Ewing, 1992a; Fayek et al., 1997a). Used nuclear fuel has lower thorium, lead, and REE concentrations (Finch and Ewing, 1992b) and therefore will likely dissolve more rapidly (USGS, 2010).

The composition and type of the secondary uranyl- mineral that will form during the alteration and dissolution of UO_2 or uraninite depends of the starting composition of UO_2 or uraninite, the composition of the fluid that is interacting with the solid phase, which is controlled largely by the host rock, and the oxidative potential of the fluid (Finch and Ewing, 1991; Finch and Ewing, 1992a,b; Fayek et al., 1997a,b; Fayek et al., 2000; Bray et al., 2007; Schindler et al., 2010; USGS, 2010). Therefore, a mineral paragenesis (a sequence of minerals precipitating in order of increasing alteration and dissolution) is generally observed (Wronkiewicz et al., 1996; Fayek et al., 2006). In general the paragenesis is uranyl oxide hydrates $\Rightarrow$uranyl silicates $\Rightarrow$

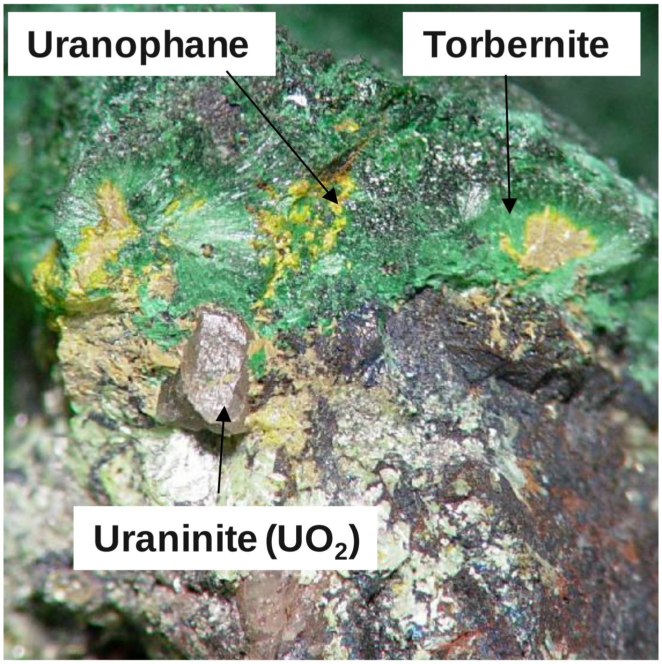


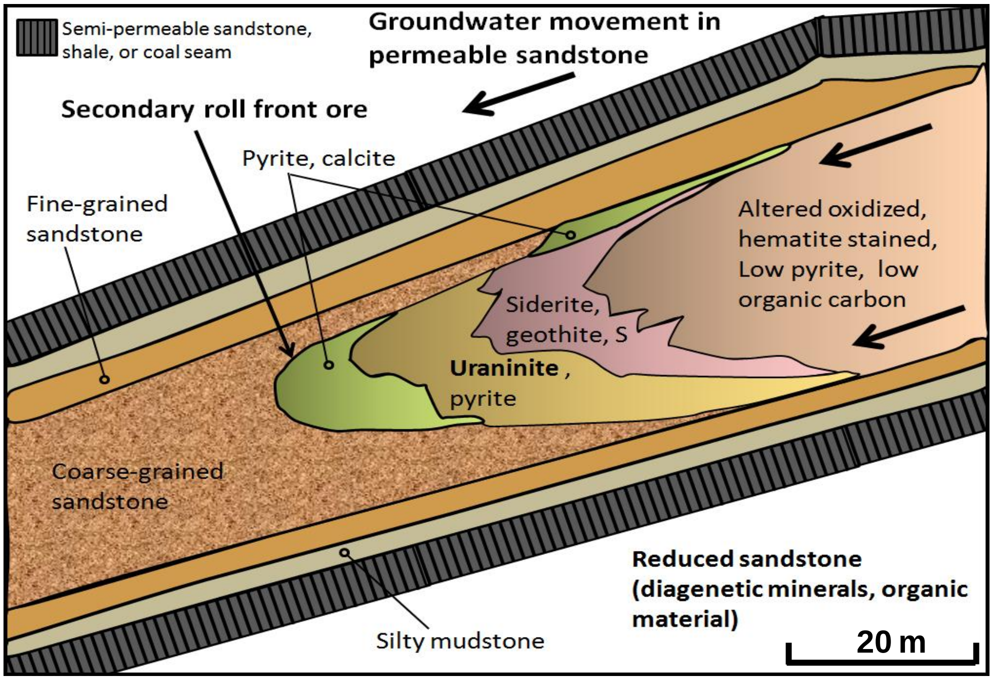
Figure 6. Uraninite is altering to secondary uranium minerals torbernite ($Cu(UO_2)_2(PO_4)_2 \cdot 12\ H_2O$) followed by uranophane ($Ca(UO_2)_2[HSiO_4]_2 \cdot 5H_2O$).

alkali- and alkaline-earth uranyl silicates + palygorskite clay (Wronkiewicz et al., 1996; USGS, 2010). Specifically, in the experimentally determined paragenetic sequence, the following minerals were identified: $UO_2 \Rightarrow$ dehydrated schoepite ($UO_3 \cdot 2H_2O$) $\Rightarrow$ compreignacite [$K_2(UO_2)_6O_4(OH)_6 \cdot 8H_2O$] + becquerelite [$Ca(UO_2)_6O_4(OH)_6 \cdot 8H_2O$] $\Rightarrow$ soddyite [$(UO_2)_2SiO_4 \cdot 2H_2O$] $\Rightarrow$ boltwoodite [$K(H_3O)(UO_2)SiO_4$] + uranophane [$Ca(UO_2)_2Si_2O_7 \cdot 6H_2O$] + palygorskite clay (Wronkiewicz et al., 1996). The experimentally-determined mineral paragenesis is nearly identical to secondary uranium phases observed during the weathering of naturally occurring uraninite under oxidizing conditions, such as that at the Nopal I uranium deposit, Peña Blanca, Mexico (Wronkiewicz et al., 1996; Fayek et al., 2006). Based on experimental data the mineral sequence appears to be controlled by precipitation kinetics. The remarkable similarity between the experimental and natural mineral paragenesis increases confidence that the experiments and the natural analogue reactions may simulate the long-term reaction progress of used UO_2 fuel.

Although the similarity between experimental data and natural analogues is promising, uraninite as an analogue for UNF must be used with care. For example, very old uraninite typically contains in-grown radiogenic lead that has accumulated through time (Finch and Ewing, 1992b). Secondary mineral formation was responsible for incorporating large quantities of uranium at Shinkolobwe, Zaire. Finch and Ewing (1991) identified 50 secondary uranium-bearing minerals. However, because the Shinkolobwe deposit is 1,800 Ma, radiogenic lead-bearing minerals formed during the alteration of uraninite. At Okélobondo, Gabon, and Shinkolobwe, other secondary minerals, such as (U,Zr)-silicates, formed stable phases (Finch and Ewing, 1991). Therefore, the uraninite used as an analogue may not be similar in composition to used nuclear fuel, which contains very little lead and may alter to form different secondary minerals (Buck et al., 1997; Corkhill et al., 2014)

For commercial nuclear reactors other than CANDU reactors, used fuel is artificially enriched in ^{235}U (e.g., 1% vs. natural abundance of 0.72% ^{235}U) and contains nuclear fission products that are generally not present in high concentrations in uraninite (USGS, 2010). In contrast, uraninite contains a higher proportion of non-radiogenic trace-element impurities. Also, the thermal history of used fuel, unlike that of uraninite, may cause lattice and structural crystallization defects in the used fuel that are not present in the uraninite (USGS, 2010). Used nuclear fuel from CANDU reactors is depleted in ^{235}U (e.g., ~0.5%) relative to natural abundance.

Nevertheless, a number of studies have relied on uranium deposits as natural analogues for processes that may occur in a DGR. Tracer studies using ^{99}Tc, ^{129}I, ^{239}Pu, and U concentrations in rocks and groundwaters surrounding Cigar Lake (Saskatchewan) and Koongarra (Australia) under reducing and oxidizing conditions, respectively, showed relatively rapid uraninite dissolution rates at Koongarra. Dissolution rates under reducing conditions at Oklo over the past approximately 2 billion years are minimal (Miller et al., 2000). Fernandez-Diaz et al. (2000) analyzed uranium isotopes in the ore and surrounding host rock at the Bangombé and Oklo uranium deposits. They concluded that very little radionuclide migration occurred over the past 2 billion years. Roll-front uranium deposits can be used as analogues to study the transport of uranium through porous rock. Roll-front deposits form in inclined, porous sedimentary (typically sandstone) host rocks, consisting of a reduced zone up dip of the ore deposit, and an oxidized zone. The uranium ore body occurs at this redox front (Fig. 7), as mineralization follows the redox-cline, separating non-oxidized from oxidized host rock as a consequence of continuously migrating and oxidizing groundwaters that dissolve and rea)



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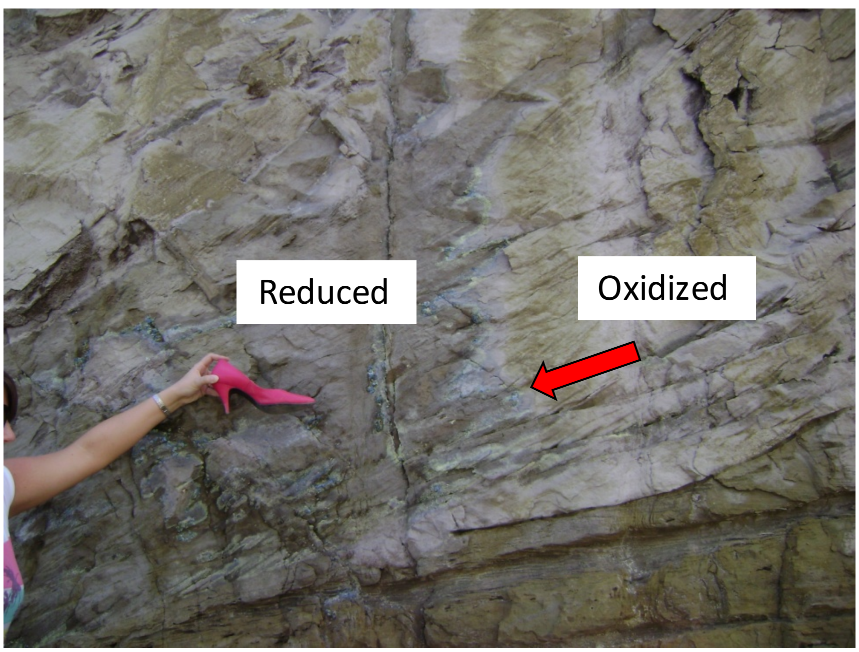

Figure 7. a) A schematic representation of a roll-front uranium deposit showing the complex structure of the ore zone, and b) an outcrop of a roll-front deposit, Turkey Creek, Colorado, USA show the reduce (grey) and oxidized zones (beige/light grey).

precipitate the uranium minerals at the redox boundary (i.e., "pushing" the ore body through the rock formation in geologic time; e.g., Osamu Utsumi mine, Brazil; Hofmann, 1999). Mineralization transects the host rock strata, creating a typically discordant, crescent shape in cross-section (Fig. 7). Uranium is reduced and precipitates as uraninite or coffinite by a variety of processes including microbial activity, and the presence of reducing carbonaceous material. Oxidized portions can extend for tens of kilometers (e.g., permeable sandstones deposited in a molasse setting in YL basin in NW China, Min et al., 2005). By investigating the mechanisms responsible for creating oxidizing or reducing conditions, we can understand how uranium and associated radionuclides in used fuel can be mobilized in a waste disposal facility, as well as the conditions that will result in their precipitation and consequent immobilization (McKee and Lush, 2004).

Radiolysis is defined as *"hydrolysis caused by radiation, resulting in production of charged species (hydrogen and hydroxide ions)"* (USGS, 2010) that can affect both the waste form and the waste package. It is uncertain whether radiolysis will be a potential problem in a DGR. Radiolysis at the time of reactor criticality at Oklo- Okélobondo mobilized a small amount of uranium, but mainly mobilized REEs from mineral core to rim (Curtis and Gancarz, 1983; Jensen and Ewing, 2001). A more recent study of radiolysis at Cigar Lake, showed that uraninite under anoxic conditions and with substantial hydrogen generation is stable over millions of years. The ore body has remained stable for billions of years, indicating the existence of processes that neutralize radiolytic oxidants because of dissolved hydrogen in groundwaters that are in contact with the ore body (Bruno and Spahiu, 2014).

The existence of natural fission reactors – such as those we now know are preserved at Oklo in Gabon, in west Africa – was predicted by Paul Kuroda (a Japanese physicist) in 1956. The Oklo phenomenon was discovered accidentally in 1972 during routine analysis of uranium ore from the site (being extracted for nuclear fuel by a French company) which showed a slightly depleted $^{235}U/^{238}U$ ratio. Criticality occurs when fissionable material can sustain a chain reaction. For criticality to occur in a uranium deposit ^{235}U would need to be sufficiently naturally enriched in addition to several other factors: a high overall concentration of uranium; a scarcity of neutron absorbers (e.g. rare earth elements); a high concentration of a moderator (such as water) and a sufficient size to sustain fission reactions. The Oklo deposits met these criteria, where would need to accumulate together with enough water to moderate the neutron energies and sustain a chain reaction. Uranium concentration and criticality can be triggered by changes in pH as well as by reducing conditions. The uranium deposits at Oklo that sustained neutron-induced fission reactions over 2 Ga, was used as a natural analogue for scenarios that might lead to nuclear criticality within and outside waste packages in the Swedish DGR concept (Oversby, 1996), which involves disposal in deep granite with chemically reducing groundwaters. The results of the study showed that the processes that occurred at Oklo to create a natural fission reactor did not impact the integrity of the orebody and in the unlikely case that criticality could be achieved in a DGR, it likely would be self-limiting and non-catastrophic (Oversby, 1996; USGS, 2010).

Although UNF will make up a large portion of the waste in a HLNW disposal site, radioactive waste may also occur in glass-form. Both natural and archeological glasses have been used as analogues for glass waste-forms. Basalt glasses are compositionally the most similar to nuclear waste glasses (Lutze et al, 1987) and have been studied under various natural environments, including ocean floor, subglacial, hydrothermal, and surface conditions (Grambow et al., 1986; Jercinovic et al., 1986; Jercinovic and Ewing, 1987; Arai et al., 1989; Cowan and Ewing, 1989). Archaeological glass artifacts that retained their colors and integrity have been found in caves and archaeological sites, dating from as far back as 3500 BCE (Yamato et al., 1992; Freestone and Gorin-Rosen, 1999; Chapman et al., 2006). Although natural and anthropogenic glasses are somewhat different in composition from borosilicate nuclear waste glass, studies of natural glass alteration indicate that glass with higher silica and alumina content and lower alkali and water content are more stable. Analogue studies have not considered radiation effects on glass over long time periods to confirm experimental results showing that radiation has little effect on glass waste stability (USGS, 2010).

2.2. Waste Package

A number of distinct waste-package designs have been developed to accommodate the different waste forms (Miller, 2000; USGS, 2010). The waste-package assembly is designed to securely contain high-level radioactive wastes, serving as the primary element in engineered barrier systems (EBS; USGS, 2010). A number of different metals are used in the manufacture of the waste package including, nickel-chromium alloys, stainless steel, iron, titanium and copper. Typically, the waste package could consist of the used fuel bundles contained within a two-part used fuel container, with an inner carbon-steel vessel and an outer corrosion-resistant copper vessel. Key concerns with respect to waste-package materials include rates of degradation and corrosion of metals caused by mechanical stress, unfavorable physical and chemical environments, microbially mediated degradation, and metallurgical factors (USGS, 2010). The long-term stability of waste package materials is directly related to the environmental conditions in the repository. Therefore, the study of metallic archeological artifacts and their preservation can provide important information of the corrosion rate of the metals to be used inwastepackage design and the conditions best suited for minimizing corrosion and degradation. Only a few of the metals being considered for waste containers can be studied using archaeological analogues (e.g., iron and steel, copper and bronze). Alloys and exotic metals such as Ti or the Ni-Cr-Mo alloys, and stainless steels (Fe-Cr-Ni alloys) cannot be studied using archaeological analogues (IAEA, 2005), though meteorites and naturally occurring metals can be used as analogues for those metals.

In the 1950s, the most northerly fortress of the Roman Empire at Inchtuthil, Scotland was excavated. A bundle of over 1 million iron nails was discovered. The outer nails were highly corroded and formed a crust that protected the inner set of nails, which showed low-levels of corrosion and degradation (Miller et al., 1994). This analogue showed that a corrosion rind can create a more reducing and protective inner environment for iron-rich waste package materials. Similarly, the Delhi iron pillar is a 1,600-year-old metal artifact (Fig. 8) showing small amounts of corrosion because of a protective rust layer coating the pillar, which consists of crystalline iron-hydrogen-phosphate hydrate and amorphous iron oxyhydroxides and magnetite (Balasubramaniam, 2000). Another iron pillar (99.7 percent iron) from Asoka, India, is constructed of welded disks and shows very little corrosion. Johnson and Francis (1980) suggested that pillar durability is a consequence of both the dry climate and the fabrication method (welding), done with minimal impurities. In summary, Johnson and Francis (1980) noted that most ancient materials are best preserved in dry environments, and that if temperatures are below the range where rapid oxidation occurs, elevated temperatures (above the dew point) are an advantage for the preservation of metals.

Copper-rich archeological artifacts, such as bronze cannons, copper, and ceremonial bronze, can also be used to estimate general corrosion rates over extended (hundreds to thousands of years) periods of time (Tylecote, 1977, 1979; Neretnieks, 1986; Miller et al., 1994; Chapman et al., 2006). These metal analogues, although differing in chemical composition from

Figure 8. The 1,600 year-old Delhi iron pillar. Image from Wikipedia. Photograph taken by Mark A. Wilson (Department of Geology, The College of Wooster).

the waste-package films, provide important insights into the way different metals survive corrosion over long time periods and illustrate the importance of passive films (USGS, 2010).

Iron meteorites may be good analogues of steel-alloy waste-package canisters. Iron meteorites are essentially nickel-iron alloys (Johnson and Francis, 1980). The study of these types of meteorites showed that the nickel content of a meteorite affects its resistance to corrosion because the better preserved minerals are those with higher chromium and nickel contents (Johnson and Francis, 1980).

A number of naturally occurring metals can be used as analogues to study the durability of metals proposed for the EBS. Awaruite (Ni_3Fe) also known as Josephinite, occurs in the nickel-iron-cobalt metal-bearing Josephine Peridotite, a serpentinized ultramafic ophiolite about 150 million years old in Oregon (Dick, 1974; Bird, 2001) consisting of andradite garnet, FeCo (wairuite), and minor to trace amounts of Ni_6Fe_4 and $CaO \cdot 2FeO$ (calciowüstite). These minerals can be used as a natural analogue for nickel-iron alloys materials that may be used in the EBS. The josephinite nuggets have alteration rims composed of Fe_3O_4 (magnetite, with maghemite) and $NiFe_2O_4$ (trevorite). Some samples of josephinite contain the high-temperature phase taenite (a disordered nickel-iron metal). The persistence of taenite for millions of years indicates that low-temperature phase change rates for taenite are exceedingly slow. The potential instability of chromium-bearing materials is illustrated by the observed natural release of chromium from chromite in the Sierra de Guanajuato ultramafic rocks (Robles-Camacho and Armienta, 2000). Corrosion appears to be concentrated along exsolution rims, analogous to structural defects on metal surfaces. Although the chromite has undergone some alteration, it has survived for more than 140 Ma. Although, the rate of corrosion for chromite may differ from those of the chromium-bearing metal alloys that may be used in the EBS, it can provide us with qualitative information such as the overall magnitude of the corrosion rate (e.g., millions of years).

Bentonite, a clay mineral, swells when wet, displays high plasticity, has a high chemical sorption capacity, and a high thermal conductivity. These properties are why most EBS incorporate bentonite as a buffer to surround canisters of used fuel. Once a breach occurs and water infiltrates the waste-package, the bentonite will swell, sealing small fractures. Its absorptive properties will cause radionuclides to move by diffusion only, thereby severely restricting radionuclide movement. Cigar Lake can be used as natural analogue for the clay mineral barrier in the EBS (Cramer, 1995). The Cigar Lake ore body is encapsulated in clays (mainly illite) that acted as a protective barrier over geologically long time scales (Cramer, 1995).

The Tsukiyoshi uranium deposit, Japan occurs within the lower part of the Mizunami Group sediments of Miocene age. These sediments contain an abundance of authigenic smectite. The amount of smectite increases generally from upper to lower horizons, and a smectite-rich encapsulates the uranium ore body. Zeolites including clinoptilolite-heulandite, mordenite, analcime, chabazite and philipsite are the second most abundant authigenic minerals. The study of these clay and zeolite rich zones showed that semctite limited the migration of uranium and as a result preserved the uranium deposit for over one million years. This smectite-zeolite envelope surrounding the Tsukiyoshi uranium deposit is regarded as a natural analogue of the buffer materials surrounding the high-level radioactive waste repository (Utada, 2003).

The Dunarobba Forest in Italy provides another good example of the effectiveness of clays in shielding material from groundwater over geological time scales (Benvegnú et al., 1988; Ambrosetti et al., 1992). This forest consists of stumps of trees that are ~2 Ma and surrounded by clay minerals. The clay minerals prevented oxygenated groundwater from reaching the wood, and maintained the appropriate geochemical conditions within the wood such that bacterial, fungal decay or chemical oxidation did not occur.

2.3. Build-out Materials

The degradation of the cementitious materials through interaction with groundwater can generate high-pH alkaline plumes that may affect the integrity of EBS materials, cause reactions with the surrounding host rock, and ultimately affect radionuclide transport (USGS, 2010). To test the durability of cements over relatively long time-scales (e.g., 1000's of years) several studies have studied the Gallo-Roman cements that are more than 1,500 years old (Thomassin and Rassineux, 1992). Calcium silicate hydrate compounds were formed early in the development of Roman cements, which used crushed vitreous fireclay as a pozzolan in the concrete mixture (Miller et al., 1994, 2000; Miller and Chapman, 1995). The calcium silicate hydrate compounds reduced the porosity and permeability of the concrete and helped to ensure its preservation for more than 2,000 years. Cementitious mortar from Hadrian's Wall (about 1,700 years old) in England (Fig. 9) has the same calcium silicate hydrate phases that are in modern Portland cement and still possesses excellent strength and stability (Miller et al., 2000). Similar results were obtained from the earliest reinforced concrete structures in Britain and in the earliest Portland cements used in sea defenses 150 years ago (Miller and Chapman, 1995).

A recent study using a 15-year old borehole located in the Tournemire Experimental Platform (Aveyron, France), evaluated the mineralogical and geochemical changes of the claystone in contact with the cementitious materials. Samples of Toarcian argillite were collected both next to and far from a CEM II cement paste and a CEM II concrete and examined using various techniques (Techer et al., 2012). They combined mineralogical and isotopic analysis of the samples and showed that the isotopic variations mimic the mineralogical changes with a somewhat extended volume of alteration. The data indicate that disturbances in a DGR will mainly affect the near field and will not extend into the geological barrier (Techer et al., 2012).

Sanidinite-facies metacarbonates (e.g., Marble Canyon and the Christmas Mountains in west Texas) can be used as natural analogues for high-silica cements that may be used in EBS. These rocks are formed as the result of high-temperature, low pressure metamorphism and metasomatism of carbonate rocks. The name is, however, somewhat of a misnomer, as they are typically composed largely of calc-silicate minerals, most of the carbonate having been lost during the metamorphism.

It is not, however, the metamorphism that formed sanidinites that is of interest when considering them as potential analogues for high-silica cements, but the processes of alteration and weathering that have occurred since their formation. They are potentially very useful analogues for high-silica cements for two reasons. First, many of the phases that occur in them are exact analogues of those in high-silica cements. Thus they are chemically, mineralogically and texturally very close to the cementitious material that may be used as build-out material in a repository. Second, because they have been exposed to weathering for reasonably long periods of time in a variety of environments, they can be used to predict how high-silica cements will respond in the repository over the mandated time periods.

While a large number of mineral phases have been found in sanidinite-facies metacarbonates, they are largely composed of calcium silicates. While the exact mineralogy varies from location to location, these rocks contain assemblages involving rankinite ($Ca_3Si_2O_7$), bredigite ($Ca_7Mg(SiO_4)_4$), larnite (□-Ca_2SiO_4), □'-Ca_2SiO_4, wollastonite ($CaSiO_3$), hatrurite (Ca_3SiO_5), tilleyite ($Ca_5Si_2O_7(CO_3)_2$), spurrite ($Ca_5Si_2O_8CO_3$) and others. Some also contain calc-magnesium silicates such as monticellite ($CaMgSiO_4$), melilites ($Ca(Mg,Al)(Si,Al)_2O_7$

Figure 9. Image of Hadrian's Wall east of Greenhead Lough, Northumberland with 1,700 year-old cementitious mortar. Image is from Wikipedia.

diopside ($CaMgSi_2O_6$) and merwinite ($Ca_3MgSi_2O_8$). Common alteration phases include: tobermorite ($Ca_5Si_6(O,OH)_{18}\cdot 5H_2O$), afwillite $Ca_3Si_2O_4(OH)_6$, foshagite, ($Ca_4Si_3O_9(OH)_2$), talc, serpentine, xonotlite $Ca_6Si_6O_{17}(OH)_2$. Geologically unusual phases, such as brownmillerite ($Ca_2(Al,Fe)_2O_5$ and mayenite ($Ca_{12}Al_{14}O_{33}$) otherwise known only from cement clinker, have also been found.

While the terminology is different, the phases that occur in these rocks are, indeed, analogous to those in high-silica cements. Classic Portland cement clinker consists of four minerals (before hydration) alite, or C_3S, which is identical to hatrurite, belite, or C_2S, which is identical to larnite, aluminate, or C_3A, and ferrite or Ca_4AF, which is identical to brownmillerite. Addition of excess silica as a binder stabilizes silicates relative to calcium hydroxide during hydration, and many of the calcium silicate hydroxides mentioned above as alteration phases in the sanidinite-facies metacarbonates are also well known in high-silica cements.

Although high-temperature sanidinite-facies metacarbonates are not geologically common, several are known. These include localities at: Crestmore, California (Eakle, 1917; Murdoch, 1955, 1961; Burnham, 1959), the Christmas Mountains aureole, Texas (Clabaugh, 1953; Joesten, 1974, 1976, 1977, 1983; Joesten and Fisher, 1988), and Marble Canyon, Texas (Bridge, 1966a,b, 1980, 1986; Anovitz and Kalin, 1990; Anovitz et al., 1991, 1992), Scawt Hill, Ireland (Tilley, 1929, 1933 1942; Tilley and Harwood, 1931), the Hatrurim formation (Mottled Zone, Israel), as part of a xenolith and autolith swarm in the Lower Zone of the Kiglapait Intrusion, Labrador (Owens, 2000); the Oslo rift, southern Norway (Goldschmidt, 1911; Tracy and Frost, 1991; Jamtveit et al., 1997), Mayen in the Laacher See region, Germany (Jasmund and Hentschel, 1964), localities in Velardena and Coahuila, Mexico (Wright, 1908; Temple and Heinrich, 1964), the Little Belt Mountains, Montana (Taylor, 1935), the Tres Hermanas Mountains (South Sister Peak), New Mexico (Homme and Rosenzweig, 1958), Carlingford, Northern Ireland (McConnell, 1954, 1955, 1960), the Isles of Skye, Much and Rhum, Scotland (Wyatt, 1953; Tilley, 1947; Hughes, 1960), Kilchoan, Ardnamurchan, Scotland (Agrell, 1965), Tokatoka, New Zealand (Mason, 1957); Lower Tunguska, Siberia (Sobolev, 1935; Reverdatto, 1964), Mysore State, India (Naidu and Covidarajulu, 1954) and the Sinai (mentioned in Gross, 1977).

The pyrometamorphism of limestone and marls at Maqarin, in Jordan, is consistent with sanidinite and pyroxene hornfels metamorphic facies, and produced cement zones that are similar to Ordinary Portland Cement. The Maqarin Natural Analogue Project investigated the evolution and persistence of the hyperalkaline groundwaters in the area, as a product of leaching from those naturally generated cement minerals (Alexander et al., 2007). The project developed a conceptual model, where the low-temperature leaching of the natural cement zones resulted in the formation of a hyperalkaline plume, is analogous to the evolution of a cementitious repository. In a review of alkaline natural analogues, Savage (2011) describes the evolution of a fluid alteration front and a mineral alteration front as the two main components associated with a high pH plume generated from fluid interaction with cement. A mineral alteration front in the host rock close to the cementitous material in the repository was preceded by the fluid alteration zone further from the repository. The hyperalkaline leachates remain highest close to the host rock / cement boundary. As alkalinity in the developing fluid front increases, the plume reacts with the host rock, alteration minerals are produced, and pH (along with Ca) falls in the migrating groundwater front as new minerals are produced (CSH close to the cement zone/repository material; further away, zeolites and/or feldspars depending on the host rock). At

Maqarin, mineral alteration consists of Calcium-Silicate-Hydrates (+/- Al) closest to the cement zone, with zeolites forming further downflow. However, the alteration pattern is complex, as with pulses of fluid flow and different episodes of mineralization and alteration, the plume continues to migrate and re-dissolve mixtures of CSH minerals and zeolites (Savage, 2011). Alexander et al. (2007) concluded that the secondary mineralization (in fractures and the host rock) reduced flow porosity, acting as seals.

2.4. Repository Infrastructure

Numerous examples in the form of caves and underground openings such as ancient mines provide evidence of the stability of natural and human made underground openings for millennia or longer (Fig. 10; USGS, 2010). Caves in many rock types, but mostly in carbonates and lava, are known to stand open for tens of thousands to millions of years in regions where no seismic activity has been documented. There are thousands of caves all over the world and many are visited by tourists (e.g. Mammoth Cave system, Kentucky, USA; Culver et al., 1999; Dom and Wicks, 2003; Woo et al., 2005; Gulden, 2009) and others are inhabited by animals (Culver et al., 1999). The ages of these caves vary from a few a million years (e.g., 4 Ma Carlsbad Caverns and 6 Ma upper level of the nearby Lechuguilla Cave; Polyak et al., 1998) to several hundred million years (345 Ma Upper Silurian Jenolan Caves, New South Wales, Australia; Osborne et al., 2006). Lava tubes have been investigated in several areas in the United States including Hawaii, New Mexico, Arizaona, Idaho and California (USGS, 2010). Only potions of these with thin roof sections have collapsed (USGS, 2010). The largest and best-studied lava tubes are in the Canary Islands. Cueva del Viento on Tenerife is over 17 km and up to 25 m wide. It has an age of 17570±150 B.P. (Carracedo et al., 2003). Caves also have been investigated in areas where strong ground motion has occurred many times (e.g., Mitchell Caverns, CA, 95 km from the epicenter of the Hector Mine earthquake (M=7.1) and 110 km from that of the Landers earthquake (M=7.3); USGS, 2010). Damage to speleothems has been widely reported, but roof collapse is rare. If general collapse has occurred in response to seismicity, it is unreported or possibly unrecognizable.

The record for anthropogenic underground openings spans a much shorter period of time than for natural openings, but it also attests to remarkable stability even in areas of strong seismicity. The Paleolithic flint mines of northern Europe and England were mined into chalk beds 3000–4000 B.C. The Egyptian tombs started approximately 1500 BCE and the Romans mined copper in 900-1800 BCE from the Orme Mine in Wales (USGS, 2010). Christians of Cappadocia, Turkey, excavated underground cities and churches during the second and 11th centuries A.D. (Toprak et al., 1994). There are thousands of tombs and tunnels in limestone and volcanic rock that are older than 2 ka and are located within areas of strong seismicity. These openings have not collapsed in more than 2,500 years (Raney, 1988).

In summary, subterranean openings seem to remain open for extended periods of time. In addition, a few archeological structures preserved in good condition above ground (such as the pyramids in Egypt or the aqueduct in Segovia, Spain) with the vast number of well-preserved subterranean openings from early recorded history and older times indicate that tunnels used in the DGR concept should serve as an effective barrier in the isolation of radioactive waste (USGS, 2010).

Figure 10. Photograph of the 4-6 Ma Carlsbad Caverns, New Mexico, USA. Image is from National Geographic's website. http://travel.nationalgeographic.com/travel/national-parks/carlsbad-caverns-national-park.

3.0 ANALOGUES FOR NATURAL BARRIERS

3.1. The Geosphere

In a DGR, natural barriers (i.e. the geosphere) provide the geological, hydrological and chemical conditions required to maintain the stability of the engineered barrier system (EBS). The geosphere additionally improves the overall safety of the deep disposal by limiting radionuclide mobility (e.g., Wikberg, 1987; Bruno et al., 1996). Therefore, nuclear waste disposal programs in many countries have used a variety of techniques and methods to study the host rock environments associated with their programs such as volcanic rocks (Yucca Mountain Project; Fayek et al., 2006; Calas et al., 2008; Schindler et al., 2010; USGS, 2010), rock salt (NEA-RWM 2013), compact argillaceous sediment (Smellie and Karlsson, 1999), and granite (Guthrie, 1991; Cramer, 1995; Min et al., 1998; Blyth et al., 2003).

To build confidence in the choice of host rock for a DGR, these countries have relied on a variety of natural analogue studies. For example, Laverov et al. (2008) studied uranium mobility associated with the Streltsovskoye uranium deposits, Russia, which are hosted in volcanic rocks. Using geochronology and mass balance calculations, they concluded that although the post-depositional fluids did interact with and alter the primary uranium ore, most of the uranium was retained in the newly formed coffinite-like amorphous U-Si phases. As a result, these phases formed an efficient geochemical barrier that prevented the long-distance migration of uranium. These data are supported by a study by Schindler et al. (2010) of the Nopal I uranium deposits, also hosted in volcanic rocks. They found that uranium was sequestered by opals forming from meteoric water that flowed along fractures and cracks with the volcanic rocks.

The Koongarra uranium Deposit, Australia, which is hosted in phyllosilicate, clay and carbonate-rich sediments, was the focus of many natural analogue studies between 1989 and 1992, which produced contradicting reports that are not fully resolved (Payne and Airey, 2006). A more recent study on the weathered and unweathered zones associated with the Koongarra deposit reported on the impact of weathering on the fate of uranium (Leijnse et al., 2001). They showed that the weathering zone has been moving downward and that groundwater velocities are highest in the weathering zone. Geochemical analysis of the weathered show that uranium concentrations do not significantly change with depth within the weathered zone, which implies that uranium is no longer moving within these zones. Therefore, movement of the weathering zone and the non-equilibrium dissolution of uranium in the ore-body play an important role in the transport of uranium (Leijnse et al., 2001).

Although there are many natural analogue studies that focus on repositories in clay and granite (e.g. Chapman, et al., 1984; Miller, et al., 1994, 2000), there are only a few published natural analogue studies that addresses repositories in salt domes (e.g. Knipping, 1989; Brenner, et al., 1999). Noseck, et al. (2008) summarizes natural analogue studies performed for high-level waste (HLW) repositories in salt in Germany. They showed that salt-domes have remained relatively intact for over 250 million years (e.g., Gorleben salt dome; Bornemann, et al. 2008). The Gorleben salt dome was also used as natural analogue to study magmatic interaction with the salt-domes and dissolution (Köthe, et al., 2007) and uplift and erosion (Jaritz, 1994; Zirngast, 1991). These studies that salt-domes are relatively stable environments that could house HLNW. However, they do note that more experimental data coupled with natural analogue information are required to build confidence in this type of DGR (Wolf et al., 2013).

3.2. Transport

3.2.1. Natural Analogues

If the canisters are breached in the repository and the radionuclides penetrate the bentonite clay buffer, radionuclides can be transported along fractures. However, minerals in fractures in the near- to far-field can provide a natural barrier that reduces radionuclide transport to the biosphere. As stated previously, the host rock may be crystalline, a compact argillaceous sediment, an anhydrite or other evaporite (Smellie and Karlsson, 1999)

In crystalline rock, radionuclide transport will likely occur along fractures. Therefore, the processes that will retard radionuclide transport include: (1) adsorption, (2) ion-exchange, and (3) precipitation or co-precipitation (mineralization). In less fractured geological systems (e.g., argillaceous sediment), matrix diffusion, molecular filtration and ion exclusion are the dominant mechanisms that may retard radionuclide transport (Smellie and Karlsson, 1999).

Multiple generations of minerals (i.e., mineral paragenesis) within faults and fractures can be analyzed using a variety of techniques to provide information on the source of fluids and amount of time the fault remained open to fluid and elemental transport. Smellie and Karlsson (1999) have reviewed the literature on radionuclide transport in mineralized fractures. In general, most studies have shown that uranium, thorium and light REEs are sequestered by iron-rich minerals, whereas heavy REEs are sequestered by carbonates.

Fracture systems associated with uranium deposits are excellent natural analogues to study radionuclide transport and retardation over geological time-scales and crustal length-scales (near and far-field processes). The Tono uranium deposit, Japan, which is hosted by Tertiary sedimentary rocks has been the subject of several analogue studies (Yoshida et al., 1994; Miller et al., 2000). It occurs within a tectonically active area and faulted rocks are common. The 3.4 km orebody was split by a fault 5-10 million years ago. This displaced a portion of the orebody 30 m upwards. Despite the tectonic activity and many nearby faults, rough mass balance calculations suggest that the majority of the uranium ore is in place and that no significant uranium transport has occurred. Therefore, this analogue shows that radionuclide mobility may be minimal in tectonically active environment provided that the appropriate redox conditions prevail. In most cases, the location for a DGR in crystalline rock will be located in a tectonically inactive region, providing even greater physical stability and security than at the Tono uranium deposit (Miller et al., 2000).

A number of natural analogue studies have investigated diffusion related transport in crystalline rock (for a summary see Smellie and Karlsson, 1999; USGS, 2010). A study of the El Berrocal granite-hosted uranium deposit, Spain, showed that the clay minerals that formed during uraninite-quartz vein emplacement acted as a natural barrier to subsequent uranium mobility (del Villar et al., 2003). Diffusion of uranium into the granite was only a few centimeters over millions of years and generally occurred along fractures, micro-fissures, and grain boundaries. It occurred with secondary minerals and was associated with relatively recent (100,000 years ago) fluid events and alteration (Heath, 1995)

Radioactive iodine is one of the potential radionuclides associated with UNF. Iodine-129 has a long half-life and understating its fate in the event that canister is breached is of great importance to DGR design concept. Loch Lomond is a land-locked lake, which was flooded by seawater between 6,900 to 5,400 years ago, resulting in a 1-m thick layer of marine clay, which bounded by freshwater sediments (Falck and Hooker, 1990; Hooker et al., 1985). The concentration of iodine in marine sediments is higher than in freshwater sediments. The concentrations of iodine diminish in the freshwater sediments with distance above and below the marine clay layer. Therefore, Loch Lomond can be used as natural analogue to study iodine diffusion in sediments, which can be used to help give confidence in saftey assessment of used fuel repositories. Although the conditions of Loch Lomond are not identical to a DGR, it serves to illustrate that under near water saturated conditions and in unconsolidated rock (sediments), relatively mobile elements such as iodine are escaping to the overlying water very slowly (thousands of years Falck and Hooker, 1990; Hooker et al., 1985).

The Oklo-Okelobondo and Bangombe natural fission reactors were used as a natural analogue to study uranium migration from the reactor zones, which contain uraninite with a similar composition to UNF (Toulhoat et al., 1996). They used uranium series and $^{235}U/^{238}U$ ratios as tracers in groundwaters. Preliminary conclusions show that there is good agreement between hydrogeology and hydrochemistry, and that at Okelobondo, uranium migration did occur but was subsequently trapped in secondary reduced zones. At Bangombe, a local organic-rich zone sufficiently buffered the redox conditions of the groundwater and reduced permeability such that the reactor zones were well preserved and there was little uranium migration into the host rock (Toulhoat et al., 1996).

Colloids are small particles that can transport radionuclides (Wieland and Spieler, 2001). Naturally occurring colloids are ubiquitous in groundwaters (e.g., colloid concentrations ranging from 0.28 to 1.35 mg/L in central Nevada; Kingston and Whitbeck, 1991). The Osamu Utsumi uranium deposit at Poços de Caldas area in Brazil has been the focus of an integrated natural analogue study. Miekeley et al. (1989, 1991a,b) studied colloidal concentrations and compositions in groundwaters associated with the Poços de Caldas area in Brazil. They showed that most of the colloids are composed of iron and organic matter, and that minor amounts of uranium are associated with these colloids. However, the study indicated that thorium and REEs are associated with the colloidal fraction. The results indicate that radionuclide and other trace-element transport by colloids is not a major factor in the geochemical processes of weathering, dissolution, and erosion of these ore deposits. In addition, colloids acts as an efficient and largely irreversible sink or trap for many elements (USGS, 2010). Therefore, iron-rich colloids that may be generated through degradation of structural steel present in EBS materials could sequester radionuclides.

Caves have also been used as natural analogues to study water seepage in underground openings. However, caves can only be used a qualitative analogues because they generally form in environments that are much different than DGRs and at much shallower depths. One of the most famous analogue sites is the limestone cave at Altamira, Spain. This cave was discovered in 1870 has well-preserved paintings that are ~14,000 years old (Valladas et al., 1992). Water seepage rates were monitored for 22 months by Villar et al., (1985). The average volume of water seeping into the cave was 7 liters per month. Although the wall and roof rock of the cave are fractured, less than 1 percent of the infiltrating water seeped into the cave. The fact that the paintings have not been bleached or dissolved near the fractures indicates that little water has seeped in along most fractures during the last 14,000 years.

Other examples of cave analogue studies include the Kartchner Caverns in Arizona (Buecher, 1999), the Mitchell Caverns, which are located on the eastern slope of the Providence Mountains in the East Mojave National Preserve, California (Stein and Warrick, 1979), Carlsbad Caverns in New Mexico (McLean, 1976; Polyak and Asmerom, 2001), and the Grotta Gigante near Trieste, Italy (Covelli et al., 1998). With the exception of Carlsbad Caverns, these studies showed that less than 2 percent of the water infiltrating the cave reached the internal chambers. At the Carlsbad Caverns, Polyak and Asmerom (2001) documented two periods of increased seepage over the last 4,000 years: one from 3,000 to 800 years B.P. and one from 440 to 290 years B.P.

A recent natural analogue study of seepage occurring in an adit at the Nopal I uranium mine in Chihuahua, Mexico, evaluated the effects of infiltration and seepage on the mobilization and transport of radionuclides (Dobson et al., 2011). This mine is hosted in welded volcanic tuffs and was used as a natural analogue site for the Yucca Mountain Project, USA. Dobson et al. (2011) mapped fractures along the roof of the adit and used a seepage collection system to collect and measure the volume of water infiltrating the fracture system. The adit was separated from surface by approximately 8 m of rock (i.e., water needed penetrate 8 m of rock to get to the collection system). A local automated weather station permitted direct correlation between local precipitation events and seepage. Dobson et al. (2011) concluded that seepage was highly heterogeneous with respect to time, location, and quantity. They produced a volume map for the amount of seepage throughout the adit. Their results showed that some zones in the back adit recorded elevated seepage volumes immediately following large (>20 mm/day) precipitation events. However, in most locations, there was a 1-6 month time lag between the rainy season and seepage, with longer times observed for the front adit. There was no correlation between fracture abundance and seepage volume. These observations indicate that even a relatively thin (8 m) rock mass can exert a noticeable damping effect on infiltration, and that flow and transport models must incorporate fracture flow heterogeneity (Dobson et al., 2011).

3.2.2. Anthropogenic Analogues

In ancient cultures where tomb preservation was important, people generally used two different methods of tomb construction. Both methods rely on the fact that water has a high surface tension, which causes preferential flow of water in small spaces relative to larger openings. The first method is the corbelled roof, which was used in tombs in Ireland, 3,100–3,200 years B.C. (Stuckless, 2000) and in Korea and adjacent China from about 277 B.C. to A.D. 668 (Lena, 2004). The tombs have vaulted ceilings with overlapping slabs of rock covered with soil. The Tomb at Newgrange, Ireland remained dry in spite of modern rainfall of more than 65 cm/yr. Some of the tombs discovered in Asia have well preserved paintings (Lena, 2004).

The second method for diverting water from tombs is referred to as a Richards' barrier (Conca et al., 1998). In this method, fine-grit material is placed over a coarse material. For example a wooden tomb or coffin is covered with a relatively impervious material such as clay, followed by a very coarse layer of boulders and gravel and finally by soil (USGS, 2010). These tombs are relatively impervious to water and the wooden coffins from ancient (1300-1500 year-old) tombs is generally well preserved (Watanabe, 1989).

4.0. COUPLED TRANSPORT PROCESSES

4.1. Hyperalkaline Fluids

There is some concern that hyperalkaline fluids could form from the interaction of water with cementitious material, which could transport radionuclides and affect the stability of EBS materials. Three natural analogues sites with hyperalkaline fluids were investigated. The Oman natural analogue site is located in the Semail Ophiolite Nappe of northern Oman. At this site alteration (serpentinization) of the ophiolite (ultramafic rock) by groundwaters generated reducing, hyperalkaline (pH = 10–12) Na-Cl-Ca-OH fluids, with associated minerals brucite [$Mg(OH)_2$] and portlandite [$Ca(OH)_2$] (Bath et al., 1987a,b; McKinley et al., 1988). These comprehensive studies examine the effects of hyperalkaline fluids on radionuclide transport processes, including solubility and speciation, colloid-transport, and microbial populations. A primary aim of the study was to test the ability of thermodynamic codes to represent and predict hyperalkaline rock/groundwater equilibrium conditions. Details of the investigations are summarized in Bath et al. (1987a) and McKinley et al. (1988).

There are two sites in Jordan (Fig. 11) that were used as natural analogues to investigate the effects of hyperalkaline fluids on radioniclide transport (Khoury and Nassir, 1982; Alexander, 1992; Alexander et al., 1992; Khoury et al., 1992; Clark et al., 1994; Alexander and Smellie, 1997; Smellie and Karlsson, 1999; USGS, 2010). The Maqarin analogue site in NW Jordan is characterized by bituminous-rich marls and a marble/cement unit formed by spontaneous *in situ* combustion of the sedimentary organic matter in sub-surface conditions. This high-temperature event produced marble and a suite of naturally formed cement minerals, including portlandite. Alteration of these units by groundwaters generated high-alkaline solutions. Water-rock interaction produce a younger Ca-K-Na-OH-SO_4 fluid and an older Ca-Na-K-OH fluid. In addition, the organic-rich marls contain a suite of trace-elements (e.g. U, Se, Ni, Ra, Sn) that are of interest for repository saftey assessments. The Maqarin analogue site is also used to study the interaction between the high-pH fluids and cements, and the transport and sinks for the trace-elements of interest. Although some secondary mineral precipitation did seal-off some flow-path surfaces, centimeter-length diffusion of trace elements into the micropores of the host rock did sequester a large portion of the trace elements (McKinley et al., 1988; Chambers et al., 1998; Smellie and Karlsson, 1999). Therefore, radionuclide diffusion will limit the mobility of radionuclides in a repository under similar conditions.

The Khushaym Matruk, Central Jordan (Fig. 11) is an analogue for intermediate to high-level nuclear waste repositories. The geology of this site is similar to the Maqarin analogue site, consisting of carbonate-rich and organic-rich rocks that have been subjected to high-temperatures (Techer et al., 2006). Secondary minerals, mainly calcite and gypsum in cracks and micro-cracks crosscut the bituminous-rich marls. Strontium isotopic data show that these minerals formed from the high-alkaline fluids generated by alteration of the cementitious phases, similar to the fluids at the Maqarin analogue site. Uranium series disequilibrium dating of calcite within these cracks indicate that fluid circulation was not continuous and occurred as several discreet events between 110,000–130,000 years (Techer et al., 2006).

Steefel and Lichtner (1998) use a multicomponent reactive transport model to investigate the coupling of mineralization and transport processes at Maqarin. The study assessed whether the observed mineral alteration is compatible with the hydrogeochemistry of the springs, and to establish the relationship between the evolution and timing of the alteration front, with the rock matrix and fracture porosities. The relative rate of secondary mineral precipitation in both the rock matrix and fracture influences the evolution of the system. When rate constants for mineral precipitation are equal, the rock matrix is cemented first and results in the downstream migration of the alteration plume. However if the rate of mineral precipitation is higher (by an order of magnitude) within the fracture, the fracture could seal first, resulting in a disruption upstream of the previously established mineral zones – similar to what is observed at Maqarin (e.g. Savage, 2011).

More recenty, Shao et al. (2013) carried out a similar modelling study that also investigated the porosity changes to the host rocks at Maqarin as a consequence of host rock alteration from high pH groundwater. Their model indicates that initial precipitation of alteration minerals occurs after several hundred years at a distance of 5 – 10 mm from the cement / analogue contact caused by the precipitation of ettringite and C-S-H minerals, similar to what was simulated by Steefel and Lichtner (1998).

4.2. Microbial Activity

Microbial populations will be present within nuclear-waste repositories. Past natural analogue and laboratory studies on microbial activity largely focused on relatively homogenous

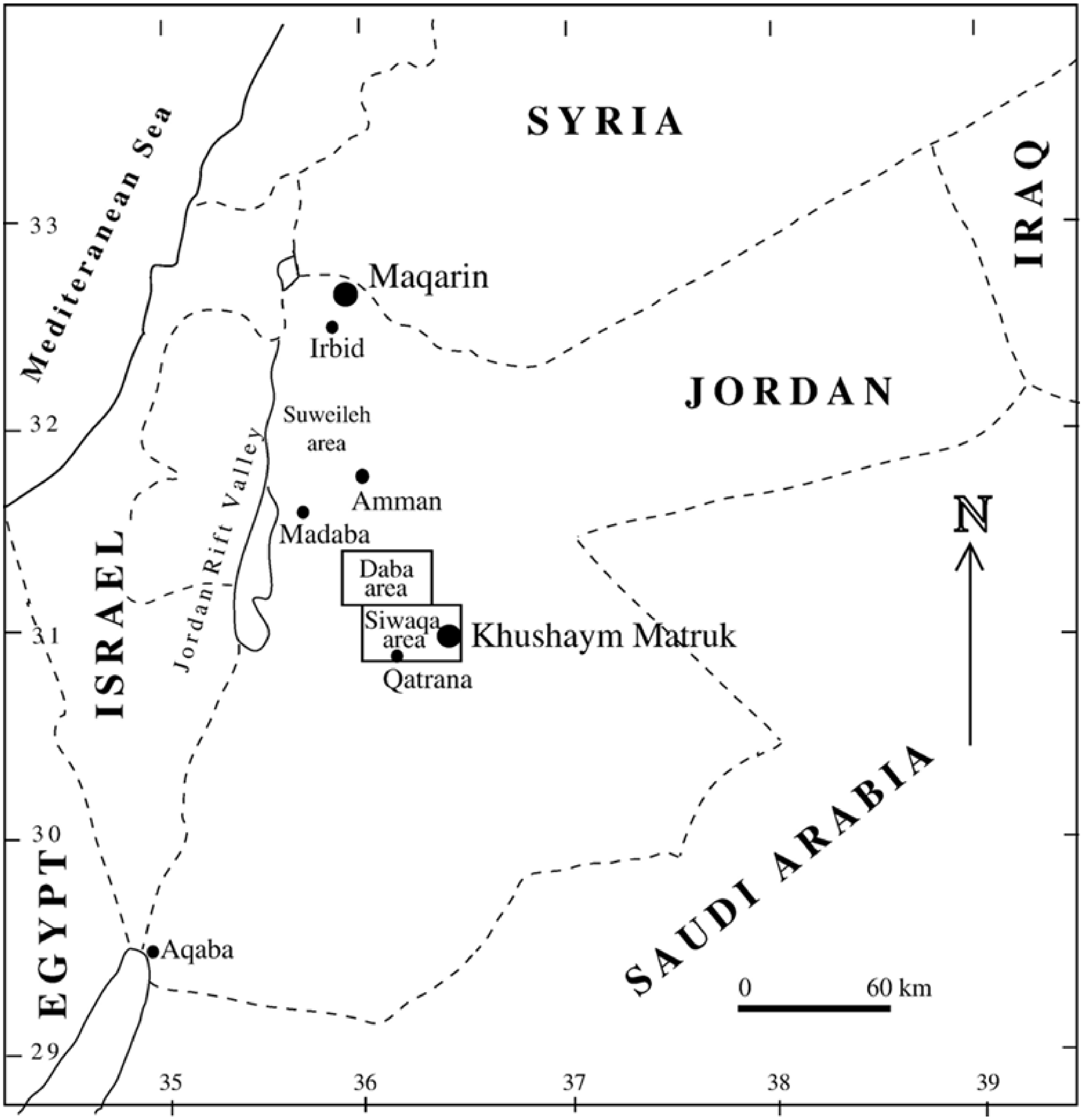

Figure 11. A map of Jordan showing the location of the Maqarin and Khushaym Matruk natural analogue sites. Image is from Techer et al. (2006).

areas within the repository (e.g., host rock or canister). However, waste repositories are complex environments that include natural and engineered barrier systems. Chemical gradients found at the contact between EBS and NB can be exploited by lithoautotrophic microbial populations (McKinley et al., 1997). Over repository time-scales (e.g., 1 million years), microbial activity will be likely be significant in areas that generate alkaline plumes and redox fonts (Williamson et al., 2014). Although a number of experiential studies (e.g., Lovley et al., 1991; Stroes-Gascoyne and West 1996; McKinley et al., 1997; Stroes-Gascoyne and West 1997; Stroes-Gascoyne et al., 1997; Libert et al., 2013) have examined microbial activity of radionuclide transport in various environments, there are few comprehensive natural analogue studies.

Natural analogue studies involving microbial activity have largely focused on granitic or crystalline bedrock environments (Pedersen and Ekendahl 1990; Jain et al., 1996; Pedersen and Haveman 1999; Haveman and Pedersen, 2002; Ahonen et al., 2004), sediments that generate alkaline fluid plumes (Nilsson, 2000; Rizoulis et al., 2014), tuffs (Kieft et al., 1997), and unconsolidated sediments (Landa, 2004; Williamson et al., 2014).

The Palmottu U–Th deposit in southwestern Finland is one of the best studied natural analogue sites for several important processes associated with HLNW disposal in crystalline rock (Miller et al., 1994). The deposit is dispersed and discontinuous and is hosted by Precambrian mica gneiss and granite (Blomqvist et al., 1998). Hydrogeochemical processes in Palmottu area include both oxidizing and reducing conditions at varying depths, which allow for the study of uranium transport at different redox potentials (Suksi et al., 1996). Iron reducing bacteria (IRB) were the dominant microbial population in Palmottu groundwater. The largest numbers of IRB were cultured from groundwaters with the lowest uranium concentrations. There was no indication of sulfur reducing bacteria (SRB). These results suggest that IRB can create a redox buffer that can affect uranium transport in solution (Haveman and Pedersen, 2002; Ahonen et al., 2004). IRB may remove uranium from solution by several mechanisms: direct enzymatic reduction of uranium; enzymatic reduction of iron, followed by chemical reduction of uranium by ferrous iron; or sorption of uranium by iron-containing minerals produced by IRB. Of these mechanisms, enzymatic U(VI) reduction by IRB is much faster than any potential non-enzymatic mechanisms (Lovley et al., 1991; Lovley and Phillips, 1992). A similar study at the Bangombe site showed similar results (Gurban et al., 1998; Haveman and Pedersen, 2002; Ahonen et al., 2004).

The Yucca Mountain Project is a DGR concept in tuffaceous volcanic rocks. Volcanic tuff was analyzed for microbial abundance and activity. Tuff was collected aseptically from nine sites along a tunnel in Yucca Mountain and samples were tested for microbial activity. In general microbial abundance was low where direct microscopic cell counts were near detection limits. Water was the major limiting factor to microbial growth and microbial activity, while the addition of nutrients such as N and P resulted in little further stimulation. Organic C stimulated growth more than water alone (Kieft et al., 1997).

Two studies of microbial activity at natural analogue sites that produce alkaline-rich fluids through interaction with natural cements have produced contradicting information. An earlier study of the Maqarin site, Jordan showed no evidence of an abundance of microbial activity associated with plume activity and Pedersen et al. (1998) suggested that groundwaters at Maqarin were too extreme for active life. However, a more recent study (Nilsson, 2000) presented preliminary results that showed that there was active uptake of carbon sources (e.g., glucose), at pH above 12. Enrichment cultures performed in Maqarin groundwater from the same sampling campaign indicated the presence of aerobic bacteria as well as anaerobic manganese, iron and sulfate-reducing bacteria.

A recent study by Rizoulis et al. (2014) investigated microbial communities and their potential for alkaline metal reduction in samples from the hyperalkaline Allas Springs, Troodos Mountains, Cyprus. The site is situated within an ophiolitic complex of ultrabasic rocks that are undergoing active low-temperature serpentinisation, which results in hyperalkaline conditions. Rizoulis et al. (2014) showed that microbial communities can exist in high pH environment, including *Hydrogenophaga* species. This indicates that alkali-tolerant hydrogen-oxidizing microorganisms could potentially colonise an alkaline geological repository. They also showed that microbial metal reduction can occur at alkaline pH. Overall, these data show that a diverse range of microbiological processes can occur in high pH environments, consistent with those expected in an intermediate level waste disposal repository.

Sediments such as uranium mine tailings are complex environments. Because of the H_2SO_4 used to extract uranium from the ores at acid-leach mills, sulfate-reducing bacteria have been identified in mine tailings (Schippers et al., 1995). Fortin and Beveridge (1997) note that sulfate reduction in sulfidic mine tailings is most likely limited by the supply of organic carbon. Nitrification and denitrification are key processes within tailings (Landa, 2004). Metals are often reduced through enzymatic processes although other processes such as chemical reduction of uranium by ferrous iron; or sorption of uranium by iron-containing minerals produced by IRB can also occur (Williamson et al., 2014).

5.0. DISCUSSION AND RECOMMENDATIONS

Several options have been proposed for the disposal of radioactive waste, including surface storage (currently being practiced at many sites), disposal in space, disposal in ice-caps, seabed disposal, and shallow land burial. However, most nations have agreed that the safest method for the disposal of intermediate and high-level nuclear waste (HLNW) is in a subsurface deep geological repository (DGR; Miller, 2000). The DGR will consist of near-field engineered barrier systems (EBS; UNF, metal canisters, clay layers etc.), deep within the geosphere (far-field). The near- and far-field barriers will be designed or selected to isolate radionuclides from the biosphere for ~1 million years. The mix of EBS and natural barriers creates a complex environment that will involve many chemical and mechanical processes that are difficult to model and predict over geological time scales (e.g., thousands or millions of years).

Therefore, one of the most challenging aspects of nuclear waste disposal in a DGR is the extrapolation of laboratory data collected over short periods of time (hours to years) to the longer periods required in saftey assessment calculations (Janeczek et al., 1996; Fayek et al., 2006). Natural and anthropogenic analogues can provide important information on the performance of waste forms (e.g., UNF) and radioactive waste repositories over geological time-scales (Janeczek et al., 1996; Fayek et al., 2006). Consequently, a there are numerous published natural analogue studies that are summarized in several books, reports and papers including McKinley (1998), Smellie and Karlsson (1999), Miller (2000); Miller, et al. (2000), West, et al. (2002), and USGS, 2010.

Using analogue studies in SA models must be done with care because there are several important limitations. For example, it may not be possible to constrain the initial conditions of a natural or anthropogenic analogue (e.g., the geosphere). The environment and materials being studied may be different from the ones considered in a repository (e.g., shallow limestone caves). Therefore, a well matched analogue for a process (e.g., coupled reactions that involve radioactivity, pressure and temperature) that may occur in a repository is difficult to find.

Although analogues can contribute valuable data to safety cases as an additional line of evidence, they cannot be relied upon as the only line of evidence.

In a recent report (NEA-RWM, 2013), Pescatore (2013) suggested that the term 'analogue' be confined to natural or anthropogenic systems that are most similar to the processes or environment in the repository that are being described. As the degree of similarity with possible repository conditions diminishes, he suggests natural and anthropogenic examples become 'analogies' or 'anecdotes' that can be used to support generic concepts associated with a DGR. He gave the following example of the nails excavated from the Roman Empire at Inchtuthil, Scotland.

"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 modeling 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 artifacts can last thousands of years underground, if it were used by a program that is not contemplating the use of steel for containers or clays as a barrier. "*

Nevertheless, natural and more importantly anthropogenic analogues can provide a visual representation of a process or concept that can help the public grasp basic rationale and principles of deep geological disposal. They represent visual examples of important functions such as long-term confinement properties of materials to illustrate potential repository situations (NEA-RWM, 2013).

5.1. General Recommendations

Although some analogue studies have provided invaluable information regarding repository selection and design, a number of analogue studies have been criticized for being too academic and failing to provide the information required for the safety case (Alexander et al., 2013). Therefore, the following are a series of recommendations:

1. Undertake a systematic review of key information, identify a set of needs (a gap analysis), and decide which of these gaps may best be fulfilled by analogue information.

2. Some researchers have suggested that national analogues (i.e. host country environments) may build more public support for a DGR especially for anthropogenic analogues (NEA-RWM, 2013). A national analogue may be more persuasive because it is geographically and culturally familiar, which may provide some reassurance to the public (NEA-RWM, 2013).

3. Develop a national analogue program that is structured and can provide input to the development of a safety case, with an emphasis on specific national geological disposal concepts.

4. Engage safety assessor early on in the process, when selecting new analogue studies or evaluating existing analogue information. This will ensure that the required information is obtained for the safety case.

5. Integrate natural analogue information with other studies (e.g., hydrogeology, rock mechanics etc.) including laboratory experiments. This will provide more quantitative data that can be used in PA models. The Maqarin study is a notable example were field studies have been integrated with laboratory experiments (e.g., sorption, column experiments; Smellie and Karlsson, 1999).

6. To build confidence in a DGR concept multiple lines of evidence are necessary. Therefore, it is recommended that an attempt is made to evaluate the true potential of analogues for public communication and dialogue through a structured opinion survey.

5.2. Scientific Recommendations

It is impossible to provide a complete list of the most critical needs for PA, which should be addressed using analogue studies because these needs will depend on specific repository designs and disposal environments. For example, large-scale analogue studies (e.g., Oklo) should be focused on environments more similar to the proposed repository site, consist of systems with relevant elements and investigate processes that directly relevant to the PA. However, it is possible to identify a few key research topics that may be relevant to several DGR concepts and that are suited to analogue studies. These are:

1. Anthropogenic analogues are deemed important for building public confidence in DGR concepts. Therefore, researchers should attempt to be part of excavations to document the preservation of the artifacts and the soil encapsulating the artifacts.

2. Analogue studies for the near-field chemical environment.

3. High pH plume site and natural cements.

4. Microbial interaction associated with high-grade uranium deposits.

5. Natural analogues for sorption and diffusion of radionuclides (e.g., long distance transport).

6. Natural analogues of secondary traps for radionuclides.

7. Colloidal transport in natural systems.

8. Natural analogues for site-specific matrix diffusion quantification.

These recommended topics can provide information that is central to the safety case or have not been completely addressed (resolved) in previous analogue studies.

6.0. REFERENCES

Agrell, S.O. (1965): Polythermal metamorphism of limestone at Kilchoan, Ardnamurchan (Scotland). *Min. Mag*., **34**, 1-15.

Ahonen, L., Kaija, J., Paananen, M., Hakkarainen, V., Ruskeeniemi, T. (2004): Palmottu natural analogue: A summary of the studies. *Geological Survey of Finland*, Report YST-121, 39p.

Alexander, W.R. (1992): A natural analogue study of cement buffered hyperalkaline groundwaters and their interaction with a sedimentary host rock, I. Source term description and geochemical database validation. *Nagra Technical Report*, NTB 91-10, Nagra Wettingen, Switzerland.

Alexander, W. R., Dayal, R., Eagleson, K., Eikenberg, J., Hamilton, E., Linklater, C.M., McKinley, G., Tweed, C.J. (1992): A natural analogue for high pH cement pore waters from the Maqarin area of northern Jordan II: results of predictive geochemical calculations. *J. Geochem. Explor*., **46**, 133-146.

Alexander, W.R., and Smellie, J.A.T. (1997): The Maqarin natural analogue project: an overview. Proc. 7th *NAWG Workshop*, Stein am Rhein, Sept. 1996. CEC EUR Report EN 18734.

Alexander, W.R., Clark, I.D., Degnan, P., Elie, M., Kamei, G., Khoury, H., Mader, U., Milodowski, A.E., Pedersen, K., Pitty, A.F., Salameh, E., Smellie, J.A.T., Techer, I., Trotignon, L. (2007): Cementitious natural analogues: Safety assessment implications for the unique systems in Jordan. Retrieved from http://www.bedrock-geosciences.com/articles.htm on July 30, 2015.

Alexander, W.R., Milodowski, A.E., Pitty, A.F., Hardie, S.M.L., Kemp, S.J., Rushton, J.C., Andreas Siathas, Avrim Siathas, Mackenzie, A.B., Korkeakoski, P., Norris, S., Sellin, P., Rigas, M. (2013): Bentonite reactivity in alkaline solutions: interim results of the Cyprus Natural Analogue Project (CNAP). *Clay Min.*, **48**, 235–249.

Ambrosetti, P., Basilici, G., Gentili, S., Biondi, E., Cerquaglia, Z., Girotti, O. (1992): La foresta fossile di Dunarobba. Ediart, Todi, Italy, 90p.

Anovitz, L.M., and Kalin, R.M. (1990): High-temperature metamorphism around the Marble Canyon intrusion, west Texas. *Geol. Soc. Amer. Abst*. **22**, A258.

Anovitz, L.M., Kalin, R.M., and Ruiz, J. (1991): High-temperature contact metamorphism: Marble Canyon west Texas. *Geol. Soc. Amer. Abst, Mineralogical Society of America Symposium*, **23**, A50.

Anovitz, L.M., Kalin, R.M., and Ruiz, J. (1992): Constraints on fluids and mass transfer during high-temperature contact metamorphism: Marble Canyon west Texas. *Geol. Soc. Amer. Abst*. **24**, A263.

Arai, T., Yusa, Y., Sasaki, N., Tsunoda, N., and Takano, H. (1989): Natural analogue study of volcanic glass - A case study of basaltic glasses in pyroclastic fall deposits of Fuji Volcano, Japan. *In*: (Lutze, W., and Ewing, R.C., Eds.,) Scientific Basis for Nuclear Waste Management XII, Berlin, Germany, October 10-13 1988 Symposium: *Materials Research Society*, **127**, 73-80.

Balasubramaniam, R. (2000): On the corrosion resistance of the Delhi iron pillar: *Corr. Sci*., **42**, 2103–2129.

Bath, A.H., Berner, U., Cave, M., McKinley, I.G., Neal, C. (1987a): Testing geochemical models in a hyperalkaline environment. *In:* (Côme, B., and Chapman, N.A., Eds.,) Natural Analogues in Radioactive Waste Disposal. Brussels, April 28-30, 1987, Symposium: *Radioactive Waste Management Series*, EUR 11037 EN, 167-178.

Bath, A.H., Christofi, N., Neal, C., Philp, J.C., Cave, M.R., McKinley, I.G., Berner, U. (1987b): Trace element and microbiological studies of alkaline groundwaters in Oman, Arabian Gulf, in A natural analogue for cement pore-waters: Technical Report 87-16, Switzerland, *Nationale Genossenschaft für die Lagerung Radioaktiver Abfalle* (*NAGRA*), 65p.

Benvegnú, F., Brondi, A., Pollizzano, C. (1988): Natural analogues and evidence of long term isolation capacity of clays occurring in Italy: contribution to the demonstration of geologic disposal reliability of long lived wastes in clay. *CEC Nuclear Science and Technology Report* EUR 11896, CEC Lexembourg.

Bird, J.M. (2001): “John M. Bird”: Ithaca, New York, Cornell University. Accessed January 30, 2015, http://www.geo.cornell.edu/geology/faculty/Bird.html

Blomqvist, R., Kaija, J., Lampinen, P., Paananen, M., Ruskeeniemi, T., Korkealaakso, J., Pitkanen, P., Ludvigson, J.-E., Smellie, J., Koskinen, L., Floria, E., Turrero, M.J., Galarza, G., Jakobsson, K., Laaksoharju, M., Casanova, J., Grundfelt, B., and Hernan, P. (1998): The Palmottu natural analogue project phase I - Hydrogeological evaluation of the site: Luxembourg, Commission of the European Communities, *Nuclear Science and Technology Report*, EUR 18202 EN, 96p.

Blyth, A., Frape, S., Ruskeeniemi, T., Blomqvist, R. (2003): Origins, closed system formation and preservation of calcites in glaciated crystalline bedrock: evidence from the Palmottu natural analogue site, Finland. *App. Geochem.*, **19**, 675–686.

Bornemann, O., Behlau, J., Fischbe ck, R., Hammer, J., Jaritz, W., Keller, S., Mingerzahn, G., Schramm, M. (2008): Description of the Gorleben Site part III: Results of the geological surface and underground exploration f the salt formation. *BGR*, Hannover, 2008, 237p.

Bray, T.H., Ling, J., Choi, E-S., Brooks, J.S., Beitz, J.V., Sykora, R.E., Haire, R.G., Stanbury, D.M., Albrecht-Schmitt, T.E. (2007): Critical role of water content in the formation and reactivity of uranium, neptunium, and plutonium iodates under hydrothermal conditions:

implications for the oxidative dissolution of spent nuclear fuel. *Inorg. Chem.*, **46**, 3663-3668.

Brenner, J., Feddersen, H-K., Gies, H., Miehe, R., Rothfuchs, T., Storck, R. (1999): Untersuchung von Altversatz als Analogon zur Konvergenz und Kompaktierung versetzter untertägiger Hohlräume im Salz über lange Zeiträume - Phase I. Gesellschaft für Anlagenund Reaktorsicherheit (*GRS*) mbH; GRS-147, 67p.

Bridge, T.E. (1966a): Contact Metamorphism in Siliceous Limestone and Dolomite in Marble Canyon and Geology of Related Intrusion, Culberson County, Trans-Pecos, Texas. Ph.d. Thesis, Univ. of Texas, Austin.

Bridge, T.E. (1966b): Bredigite, larnite, and dicalcium silicates from Marble Canyon. *Am. Min*., **51**, 1766-1774.

Bridge, T.E. (1980): Progressive metamorphism of Permian siliceous limestone and dolomite - A complete sequence around a monzonite intrusion, Marble Canyon, Diablo Plateau, Texas. *New Mexico Geol. Soc. Guidebook*, 31st Field Conference, Trans-Pecos Region, 225-229.

Bridge, T.E. (1986): Contact metamorphism around the Marble Canyon stock. *In*: (Price, J.G., Henry, C.D., Parker, D.F., Barker, D.S., Eds.,) Igneous Geology of Trans-Pecos, Texas: Field Trip Guide and Research Articles. *Univ. Texas Bur. Econ. Geol*. Guidebook, **23**, 26-30.

Bruno, J., Cera, E., Duro, L., Ahonen, L. (1996): Deep groundwater redox reactions in the Palmottu uranium deposit: The role of uranium and iron in these processes. *Geological Survey of Finland*, Posiva Report 96-24, 26p.

Bruno, J., and Ewing, R. (2006): Spent Nuclear Fuel. *Elements*, **2**, 343-349.

Bruno, J., and Spahiu, K. (2014): The long-term effect of hydrogen on the UO_2 spent fuel stability under anoxic conditions: Findings from the Cigar Lake Natural Analogue study. *App. Geochem.*, **49**, 178-183.

Buck, E.C., Wronkiewicz, D.J., Finn, P.A., Bates, J.K. (1997): A new uranyl oxide hydrate phase derived from spent fuel alteration. *J. Nuc. Mater.*, **249**, 70-76.

Buecher, R.H. (1999): Microclimate study of Kartchner Caverns, Arizona. *J. Cave Karst Stud.*, **61**, 108-120.

Burnham, C.W. (1959): Contact metamorphism of magnesian limestones at Crestmore, California. *Bull. Geol. Soc. Am*., **70**, 879-920.

Calas, G., Agrinier, P., Allard, T., Ildefonse, P. (2008): Alteration geochemistry of the Nopal I uranium deposit (Sierra Pena Blanca, Mexico), a natural analogue for a radioactive waste repository in volcanic tuffs. *Terra Nova*, **20**, 206-212.

Carracedo, J.C., Paterne, M., Guillou, H., Perez Torrado, F.J., Paris, R., Badiola, E.R., Hansen, A. (2003): Dataciones radiometricas (14C y K/Ar) del Teide y el Rift NO, Tenerife, Islas Canarias. *Estudios Geologicos*, **59**, 15-29.

Chambers, A.V., Haworth, A., Ilett, D., Linklater, C.M., Tweed, C.J. (1998): Geochemical modelling of hyperalkaline water/rock interactions. *In*: (Smellie, J.A.T., Ed.,) Maqarin Natural Analogue Study, Phase III, volume I: Svensk Kärnbränsleförsörjning, *SKB* TR-98-04, **1**, 401p.

Chapman, N.A., McKinley, I.G., Smellie, J.A.T. (1984): The potential of natural analogues in assessing systems for deep disposal of high-level radioactive waste: Swedish Nuclear Fuel and Waste Management Company, *SKB Technical Report* 84-16 103p.

Chapman, N.A., and Smellie, J.A.T. (1986): Introduction and summary of the workshop. *Chem. Geol.*, **55**, 167-173.

Chapman, N., Dansie, A., McCombie, C. (2006): Preservation of archaeological material in arid environments relevant to Yucca Mountain. Proceedings of the 11th International High-Level Radioactive Waste Management Conference, Las Vegas, Nevada, April 30–May 4, 2006: *American Nuclear Society*, La Grange Park, Illinois, 691-697.

Clabaugh, S. (1953): Contact metamorphism in the Christmas Mountains, Brewster county, Texas. *Bull. Geol. Soc. Amer*., **64**, 1408.

Clark, I.D., Dayal, R., Khoury, H.N. (1994): The Maqarin (Jordan) natural analogue for 14-C attenuation in cementitious barriers. *Waste Manag*., **14**, 467-477.

CNSC (2006): Assessing the long-term safety of radioactive waste management/Regulatory Guide G–320

Conca, J.L., Apted, M.J., Zhou, W., Arthur, R.C., Kessler, J.H. (1998): Flow barrier system for the long-term high-level-waste isolation - Experimental results. *Nucl. Tech.*, **1124**, 88-100.

Corkhill, C.L., Myllykylä, E., Bailey, D.J., Thornber, S.M., Qi, J., Maldonado, P., Stennett, M.C., Hamilton, A., Hyatt, N.C. (2014): Contribution of energetically reactive surface features to the dissolution of CeO_2 and ThO_2 analogues for spent nuclear fuel microstructures. *App. Mater. Inter.*, **6**, 12279-12289.

Covelli, S., Cucchi, F., Mosca, R. (1998): Monitoring of percolation water to discriminate surficial inputs in a karst aquifer. *Environ. Geol.*, **36**, 296-304.

Cowan, R., and Ewing, R.C. (1989): Freshwater alteration of basaltic glass, Hanauma Bay, Oahu, Hawaii - A natural analogue for the alteration of borosilicate glass in freshwater. *In:* (Lutze, W., and Ewing, R.C., Eds.,) Scientific Basis for Nuclear Waste Management XII, Symposium, Berlin, Germany, October 10-13, 1988: *Mater. Resear. Soc.*, **127**, 49-56.

Cramer, J.J. (1995): The Cigar Lake uranium deposit - Analog information for Canada's nuclear fuel waste disposal concept: *Atomic Energy Canada Limited*, AECL-11204, 32p.

Culver, D.C., Hobbs III, H.H., Christman, M.C., Master, L.L. (1999): Distribution map of caves and cave animals in the United States. *J. Cave Karst Stud.*, **61**, 139-140.

Curtis, D.B., and Gancarz, A.J. (1983): Radiolysis in nature - Evidence from the Oklo natural reactors. Swedish Nuclear Fuel and Waste Management Company, *SKB Technical Report* 83-10, 60p.

Dick, H.J.B. (1974): Terrestrial nickel-iron from the Josephine peridotite - Its geologic occurrence, associations, and origin. *Earth Planet. Sci. Lett.*, **24**, 291-298.

Dobson, P.F., Ghezzehei, T.A., Cook, P.J., Rodriguez-Pineda, J.A., Villalba, L., de la Garza, R. (2011): Heterogeneous seepage at the Nopal I natural analogue site, Chihuahua, Mexico. *Hydrogeol. J*. **20**, 155–166.

Dom, J.E., and Wicks, C.M. (2003): Morphology of the caves of Missouri. *J. Caves Karst Stud.*, **66**, 155-159.

Eakle, A.S. (1917): Minerals associated with the crystalline limestone at Crestmore, Riverside County, California. *Univ. Calif. Bull. Dept. Geol*., **10**, 327-360.

Ewing, R.C. (1999): Nuclear waste forms for actinides. *Proc. Natl. Acad. Sci. USA*, **96**, 3432-3439.

Falck, W.E., and Hooker, P.J. (1990): Quantitative interpretation of Cl, Br, and I pore water concentration profiles in lake sediments of Loch Lomond, Scotland. *BGS Technical Report*, WE/90/3, 49p.

Fayek, M., Janeczek, J., Ewing, R.C. (1997a): Mineral chemistry and oxygen isotopic analyses of uraninite, pitchblende and uranium alteration minerals from the Cigar Lake deposit, Saskatchewan, Canada. *App. Geochem.*, **12**, 549-565.

Fayek, M., Kyser, T.K., Ewing, R.C., Miller, M.L. (1997b): Uraninite-water interaction in an oxidizing environment. *Mat. Res. Soc. Symp. Proc.*, **465**, 1201-1208.

Fayek, M., Harrison, T.M., Grove, M., Coath, C.D. (2000): A rapid in situ method for determining the ages of uranium oxide minerals: evolution of the Cigar Lake deposit, Athabasca Basin. *Int. Geol. Rev*., **42**, 163-171.

Fayek, M., Ren, M., Goodell, P., Dobson, P., Saucedo, A., Kelts, A., Utsunomiya, S., Ewing, R.C., Riciputi, L.R., Reyes, I. (2006): Paragenesis and geochronology of the Nopal I uranium deposit, Mexico. Proceedings of the 2006 International High-Level Radioactive Waste Management Conference, April 30-May 4, 2006, Las Vegas, Nevada: **American Nuclear Society**, La Grange Park, Illinois, 55-63.

Fernandez-Diaz, M., Quejido, A.J., Crespo, M.T., Perez del Villar, L., Martin-Sanchez, A., Lozano, J.C. (2000): Uranium isotopic data in uraninite spent fuel from the Bangombe natural nuclear reactor (Gabon) and its Surroundings. *App. Rad. Iso.*, **53**, 91-96.

Finch, R.J., and Ewing, R.C. (1991): Alteration of natural UO_2 under oxidizing conditions from Shinkolobwe, Katanga, Zaire - A natural analogue for the corrosion of spent fuel. *Radiochim. Acta*, **52-53**, 395-401.

Finch, R.J., and Ewing, R.C. (1992a): The corrosion of uraninite under oxidizing conditions. *J Nucl. Mater*., **190**, 133-156.

Finch, R.J., and Ewing, R.C. (1992b): Alteration of natural uranyl oxide hydrates in Si-rich groundwaters - Implications for uranium solubility. *In*: (Sombret, C.G., Ed.,) Scientific Basis for Nuclear Waste Management XV, Symposium, Strasbourg, France, November 4-7, 1991. *Mater. Res. Soc.*, **257**, 465-472.

Fortin, D., and Beveridge, T.J. (1997): Microbial sulfate reduction within sulfidic mine tailings: formation of diagenetic Fe sulfides. *Geomicrobio. J.*, **14**, 1-21.

Freestone, I.C., and Gorin-Rosen, Y. (1999): The great glass slab of Bet She'arim, Israel - An early Islamic glassmaking experiment? *J. Glass Stud.*, **41**, 105-116.

Goldschmidt, V. (1911): The Nature of crystals. *Science*, **33**, p.871.

Grambow, B., Jercinovic, M.J., Ewing, R.C., Byers, C.D. (1986): Weathered basalt glass - A natural analogue for the effects of reaction progress on nuclear waste glass alteration. *In:* (Werme, L.O., Ed.,) Scientific Basis for Nuclear Waste Management IX, Symposium, Stockholm, Sweden, September 9-11, 1985. *Mater. Res. Soc.*, **50**, 263-272.

Gross, S. (1977): The mineralogy of the Hatrurim formation, Israel. *Israel Geol. Surv. Bull.* ***70***, 80p.

Gulden, B. (2009): The NSS Geo2 long & deep caves Web site: http://www.caverbob.com, accessed January 10, 2015.

Gurban, I., Laaksoharju, M., Ledoux, E., Made, B., Salignac, A.L. (1998): Indications of uranium transport around the reactor zone at Bangombé (Oklo). Swedish Nuclear Fuel and Waste Management Company, *SKB Technical Report* 98-06, 30p.

Guthrie, V. (1991): Determination of recent ^{238}U, ^{234}U and ^{230}Th mobility in granitic rocks: application of a natural analogue to the high-level waste repository environment. *App. Geochem.*, **6**, 63-74.

Haveman, S.A., and Pedersen, K. (2002): Microbially mediated redox processes in natural analogues for radioactive waste. *J. Contam. Hydrol.*, **55**, 161– 174.

Heath, M.J. (1995): Rock matrix diffusion as a mechanism for radionuclide retardation - Natural radioelement migration in relation to the microfractography and petrophysics of fractured crystalline rock. Luxembourg, *Commission of the European Communities*, EUR 15977 EN, 128p.

Hofmann, B. A. (1999): Geochemistry of Natural Redox Fronts - A Review. *NTB* 99–05, Wettingen, Switzerland: NAGRA.

Homme, F.C., and Rosenzweig, A. (1958): Spurrite and monticellite skarns in the Tres Hermanas Mountains, Luna County, New Mexico. *Bull. Geol. Soc. Amer*., **69**, p.1586.

Hooker, P. J., MacKenzie, A. B., Scott, R. D., Ridgway, I. M., McKinley, I. G., West, J. M. (1985): A study of natural and long term (10^3 - 10^4y) elemental migration in saturated clays and sediments, Part III. *British Geological Survey Report*, FLPU 85-9.

Hughes, C.J. (1960): An occurrence of tilleyite-bearing limestones in the Isle Rhum, Inner Hebrides. *Geol. Mag*., **97**, 384-388.

IAEA (1989): Natural analogues in performance assessment for the disposal of radioactive waste. *IAEA Technical Report* 304p.

IAEA (1999): Use of natural analogues to support radionuclide transport models for deep geological repositories for long lived radioactive wastes. TECDOC-1109, 39p.

IAEA (2005): Anthropogenic analogues for geological disposal of high level and long lived waste. TECDOC-1481, 73p.

IAEA (2012): Standards: The Safety Case and Safety Assessment for the Disposal of Radioactive Waste. *Specific Safety Guide* No. SSG-23, 120p.

Jain, D.K., Stroes-Gascoyne, S., Providenti, M., Tanner, C., Cord, I. (1996): Characterization of microbial communities in deep groundwater from granitic rock. *Can. J. Microbial*., **43**, 272-283.

Jamtveit B., Dahlgren, S. Austrheim H. (1997): High-grade contact metamorphism of calcareous rocks from the Oslo Rift, Southern Norway. *Am. Min.*, **82**, 1241-1254.

Janeczek, J., and Ewing, R.C. (1992): Dissolution and alteration of uraninite under reducing conditions: *J. Nucl. Mater.*, **190**, 157-173.

Janeczek, J., Ewing, R.C, Oversby, V.M., Werme, L.O. (1996): Uraninite and UO_2 in spent nuclear fuel: a comparison. *J. Nucl. Mater.*, **238**, 121-130.

Jaritz, W. (1994): Die Entwicklungsgeschichte des Strandortes Gorleben als naturliches analogon fur das langzeitverhalten eines barrierensstems. *Z. dt. geol. Ges*., **145**, 192-206.

Jasmund K, and Hentschel, J. (1964): Seltene Mineralparagenesen in den Kalksteineinschlüssen der Lava des Ettringer Bellerberges bei Mayen (Eifel). *Beitr. Mineral. Petrogr.*, **10**, 296–314.

Jensen, K.A., and Ewing, R.C. (2001): The Okélobondo natural fission reactor, southeast Gabon –geology, mineralogy, and retardation of nuclear-reaction products: *Geol. Soc. Am. Bull.*, **113**, 32-62.

Jercinovic, M.J., Ewing, R.C., Byers, C.D. (1986): Alteration products of basalt glass from Frenchman Springs flow, Wanapum Basalts, Hanford, Washington. *In:* (Clark, D.E., White, W.B., Machiels, A.J., Eds.,) Nuclear Waste Management II, *Third Annual Symposium on Ceramics in Nuclear Waste Management*, Chicago, Illinois, April 28-30, 1986, Advances in Ceramics, **20**, 671-679.

Jercinovic, M.J., and Ewing, R.C. (1987) Basaltic glasses from Iceland and the deep sea – natural analogues to borosilicate nuclear waste-form glass. *JSS Project Technical Report* 88-01, Swedish Nuclear Fuel and Waste Management Company, 211 p.

Joesten, R. (1974): Local equilibrium and metasomatic growth of zoned calc-silicate nodules from a contact aureole, Christmas Mountains, Big Bend region, Texas. *Am. J. Sci*., **274**, 876-901.

Joesten, R. (1976): High-temperature contact metamorphism of carbonate rocks in a shallow crustal environment, Christmas Mountains, Big Bend Region, Texas. *Amer. Mineral*., **61**, 776-781.

Joesten, R. (1977): Mineralogical and chemical evolution of contaminated igneous rocks at a gabbro-limestone contact, Christmas Mountains, Big Bend Region, Texas. *Geol. Soc. Am. Bull*. **88**, 1515-1529.

Joesten, R. (1983): Grain growth and grain boundary diffusion in quartz from the Christmas Mountains (Texas) contact aureole. *Am. J. Sci*., **283-A** (Orville Vol.), 233-254.

Joesten, R., and Fisher, G. (1988): Kinetics of diffusion controlled mineral growth in the Christmas Mountains (Texas) contact aureole. *Geol. Soc. Am. Bull*., **100**, 714-732.

Johnson, A.B., Jr., and Francis, B. (1980): Durability of metals from archaeological objects, metal meteorites, and native metals. *Pacific Northwest Laboratory*, PNL-3198.

Khoury, H., and Nassir, S. (1982): High temperature mineralization in the bituminous limestone in Maqarin area, North Jordan. *N. Jhb. Mineral. Abh*., **144**, 197-213.

Khoury, H.N., Salameh, E., Clark, I.D., Fritz, P., Milodowski, A.E., Cave, M.R., Bajjali, W., Alexander, W.R. (1992): A natural analogue of high pH cement pore waters from the Maqarin area of northern Jordan-I, Introduction to the site. *J. Geochem. Explor.*, **46**, 117-132.

Kieft, T.L., Kovacik Jr., W.P., Ringelberg, D.B., White, D.C., Haldeman, D.L., Amy, P.S., Hersman, L.E. (1997): Factors Limiting Microbial Growth and Activity at a Proposed High Level Nuclear Repository, Yucca Mountain, Nevada. *App. Environ. Microbio.*, **63**, 3128-3133.

Kingston, W.L., and Whitbeck, M. (1991): Characterization of colloids found in various groundwater environments in central and southern Nevada. *Department of Energy*, DOE/NV/10384-36, Las Vegas, Nevada, Desert Research Institute, Water Resources Center, 136 p.

Knipping, B.J. (1989): Basalt intrusions in evaporites*: Lect. Notes Earth Sci.*, **24**, Berlin, Springer, 131p.

Köthe, A., Hoffmann, N., Krull, P., Zwirner, R. (2007) Die geologie des deckgebirges und strukturgeologie des salzstocks Gorleben. *Geol. Jahrbuch, Reihe C*, **72**, 201p.

Kotzer, T. G., and Kyser, T. K. (1995): Petrogenesis of the Proterozoic Athabasca Basin, northern Saskatchewan, Canada, and its relation to diagenesis, hydrothermal uranium mineralization and paleohydrogeology. *Chem. Geol.*, **120**, 45–89.

Laverov, N.P., Petrov, V.A., Poluektov, V.V., Nasimov, R.M., Hammer, J., Burmistrov, A.A., Shchukin, S.I. (2008): The Antei Uranium Deposit: A Natural Analogue of an SNF Repository and an Underground Geodynamic Laboratory in Granite. *Geol. Ore Dep.*, **50**, 339-361.

Leijnse, A., van de Weerd, H., Hassanizadeh, S.M. (2001): Modeling Uranium Transport in Koongarra, Australia: The Effect of a Moving Weathering Zone. *Math. Geol.*, **33**, 1-29.

Lena, K. (2004): Koguryo tomb murals: Seoul, ICOMOS-Korea and Cultural Properties Administration, 108 p.

Libert, M., Schütz, M.K., Esnault, L., Féron, D., Bildstein, O. (2013): Impact of microbial activity on the radioactive waste disposal: long term prediction of biocorrosion processes. *Bioelectrochem.*, http://dx.doi.org/10.1016/j.bioelechem.2013.10.001.

Lopez, C.R., Rodriguez, J., Recreo, F., Ruiz, C., Prado, P., Gimeno, M.J., Auque, L.F., Gomez, J., Acero, P., Gonzalez, A., Samper, J., Montenegro, L., Molinero, J., Delgato, J., Criado, A., Martinez, J.A., Ruiz, S. (2004): Analogue application to safety assessment and communication of radioactive waste geologic disposal – illustrative synthesis. *Consejo Seguridad Nuclear*, 167p.

Lovley, D.R., and Phillips, E.J.P. (1992): Reduction of uranium by Desulfovibrio desulfuricans. *Appl. Environ. Microbiol*., **58**, 850–856.

Lovley, D.R., Phillips, E.J.P., Gorby, Y.A., Landa, E.R. (1991): Microbial reduction of uranium. *Nature*, **350**, 413-416.

Lutze, W., Grambow, B., Ewing, R.C., Jercinovic, M.J. (1987): The use of natural analogues in the long-term extrapolation of glass corrosion processes. *In:* (Côme, B., and Chapman, N.A., Eds.,) Natural Analogues in Radioactive Waste Disposal – Symposium, Brussels, April 28-30, 1987. *Radioactive Waste Management Series*, EUR 11037 EN, 142-152.

Mason, B. (1957): Larnite, scawtite and hydrogrossular from Tokatoka, New Zealand. *Am. Min*., **42**, 379-392.

McConnell, J.D.C. (1954): The hydrated calcium silicates riversideite, tobermorite and plombierite. *Min. Mag*., **30**, 293-305.

McConnell, J.D.C. (1955): The hydration of larnite (β-Ca_2SiO_4) and bredigite (α-Ca_2SiO_4) and the properties of the resulting gelatinous mineral plomierite. *Min. Mag.*, **30**, 672-680.

McConnell, J.D.C. (1960): Vaterite from Ballycraigy, larne, Northern Ireland. *Min. Mag.*, **32**, 534-544.

McKee, P., and Lush, D. (2004): Natural and anthropogenic analogues: insights for management of spent fuel. *NWMO* background paper, 631-22904.101, 33p.

McKinley, I.G., Hagenlocher, I., Alexander, W.R., Schwyn, B. (1997): Microbiology in nuclear waste disposal: interfaces and reaction fronts. *FEMS Microbio. Rev.*, **20**, 545-556.

McKinley, I.G., Bath, A.H., Berner, U., Cave, M., Neal, C. (1988): Results of the Oman analogue study. *Radiochim. Acta,* **44-45**, 311-316.

McLean, J.S. (1976): Factors altering the microclimate in Carlsbad Caverns, New Mexico. *U.S. Geological Survey* Open-File Report 76-171, 56p.

Miekeley, N., Coutinho de Jesus, H., Porto da Silveira, C.L., Kuechler, I.L. (1989): Colloid investigations in the Poços de Caldas natural analogue project. *In:* (Lutze, W., and Ewing, R.C., Eds.,) Scientific Basis for Nuclear Waste Management XII, Symposium, Berlin, Germany, October 10-13, 1988. *Mater. Res. Soc.*, **127**, 831-842.

Miekeley, N., Coutinho de Jesus, H., Porto da Silveira, C.L., Linsalata, P., Morse, R., Osmond, J.K. (1991a): Natural series radionuclide and rare-earth element geochemistry of waters from the Osamu Utsumi Mine and Morro do Ferro analogue study sites, Poços de Caldas, Brazil, Stockholm, Sweden, Swedish Nuclear Fuel and Waste Management Company, *SKB Technical Report* 90-17, 70p.

Miekeley, N., Coutinho de Jesus, H., Porto da Silveira, C.L., and Degueldre, C. (1991b): Chemical and physical characterisation of suspended particles and colloids in waters from the Osamu Utsumi Mine and Morro do Ferro analogue study sites, Poços de Caldas, Brazil. *SKB Technical Report* 90-18, Stockholm, Sweden, Swedish Nuclear Fuel and Waste Management Company, 78p.

Miller, W.M. (1996): Archaeological materials as natural analogues for the geological disposal of radioactive waste. *In*: (Corfield, H.P., Nixon, T., Pollard, M., Eds.,) Preserving Archaeological Remains In Situ – Conf. Prec. Museum of London.

Miller, W. (2000): Analogues of Repository Materials, In: Geological Disposal of Radioactive Wastes and Natural Analogues: Lessons from Nature and Archaeology (Ed. W. Miller). EBSCO Publishing, 310p.

Miller, W., and Chapman, N. (1995): Postcards from the past – archaeological and industrial analogues for deep repository materials: *Radwaste Mag.*, **2**, 32-42.

Miller, W., Alexander, R., Chapman, N., McKinley, I., Smellie, J. (1994): Natural analogue studies in the geological disposal of radioactive wastes – studies in environmental science, **57**, New York, Elsevier, 395 p.

Miller, W., Alexander, R., Chapman, N., McKinley, I., Smellie, J. (2000): Geologic disposal of radioactive wastes and natural analogues – lessons from nature and archaeology. *Waste Management Series*, New York, Pergamon, **2**, 316 p.

Miller, W., Hooker, P., Smellie, J.A.T., Dalton, J., Degnan, P., Knight, P., Nosek, U., Ahonen, L., Laciok, A., Trotignon, L., Wouter, L., Hernan, P., Vela, A.L. (2006): NAnet Project (Network to review natural analogue studies and their application to repository safety assessment and public communication) Synthesis Report: European Community contract no. FIKW-CT-2002-20204, 67p.

Min, M.Z., Zhai, J.P., Fang, C.Q. (1998): Uranium-series radionuclide and element migration around the Sanerliu granite-hosted uranium deposit in southern China as a natural analogue for high-level radwaste repositories. *Chem. Geol.*, **144**, 313-328.

Min, M., Fang, C., Chen, J., Fayek, M. (2005): Petrography and genetic history of coffinite and uraninite from the Liueryiqi granite-hosted uranium deposit, SW China. *Ore Geol. Rev.*, **26**, 198-206

Murdoch, J. (1955): Bultfonteinite from Crestmore, California. *Am. Min.*, **40**, 900-906.

Murdoch, J. (1961): Crestmore, past and present. *Am. Min.*, **46**, 245-257.

Naidu, M.G., and Covidarajulu, B.V. (1954): Occurrence of calciphyres near Mallarajanahundi, Nanjangud (Mysore State). *Current Sci*., **23**, p10.

NAWG website: http://www.natural-analogues.com, accessed on 02/13/2015.

National Geographic website: http://travel.nationalgeographic.com/travel/national-parks/carlsbad-caverns-national-park, accessed 03/12/2015.

NEA-RWM (2013): Local communities' expectations and demands on monitoring and the preservation of records, knowledge and memory of a deep geological repository. NEA/RWM/R(2013)4/PROV, 66p.

Neretnieks, I. (1986): Investigations on old bronze cannons. *In*: (Côme, B., and Chapman, N.A., Eds.,) Commission of European Communities natural analogue working group, Second Meeting, Interlaken, Switzerland, June 17-19, 1986: Luxembourg, Commission of the European Communities, EUR 10671 EN, 191-197.

Nilsson, E.L. (2000): Microbes in the hyper alkaline springs of Maqarin, Jordan, a natural analogue for water/cement interaction. *Goldschmidt 2000, Journal of Conference Abstracts*, **5**, p.746.

Noseck, U., Müller-Lyda, I., Steininger, W. (2008): "Overview on Natural Analogue Studies in Germany", *ATW*, **53**, 342-348.

Osborne, R.A.L., Zwingmann, H., Pogson, R.E., Colchester, D.D.M. (2006): Carboniferous clay deposits from Jenolan caves, New South Wales – implications for timing of speleogenesis and regional geology. *Australian J. Earth Sci.*, **5**, 377-405.

Oversby, V.M. (1996): Criticality in a high-level waste repository – a review of some important factors and an assessment of the lessons that can be learned from the Oklo reactors. Svensk Kärnbränsleförsörjning, *SKB* TR-96-07, 41p.

Owens, B.E. (2000): High-temperature contact metamorphism of calc-silicate xenoliths in the Kiglapait Intrusion, Labrador. *Am. Min*., **85**, 1595-1605.

Payne, T.E., and Airey, P.L. (2006): Radionuclide migration at the Koongarra uranium deposit, Northern Australia – Lessons from the Alligator Rivers analogue project. *Phys. Chem. Earth*, **31**, 572-586.

Pedersen, K., Arlinger, J., Erlandsson, A., Hallbeck, L. (1998): *In*: (Smellie, J.A.T., Ed.) Maqarin Natural Analogue Study: Phase III. *SKB Technical Report*, TR-98-04, **1**, 367-387.

Pedersen, K., and Ekendahl, S. (1990): Distribution and activity of bacteria in deep granitic groundwaters of Southeastern Sweden. *Microb. Ecol.*, **20**, 37-52.

Pedersen, K., and Haveman, S. (1999): Analysis of diversity and distribution of microorganisms in Palmottu groundwater and evaluation of their influence on redox potential and uranium (VI) reduction. *Geol. Surv. Finland*, Technical Report 99-14, 12p.

Pérez del Villar, L., Reyes, E., Delgado, A., Núñez, R., Pelayo, M. and Cózar, J.S. (2003): Argillization processes at the El Berrocal analogue granitic system (Spain): mineralogy, isotopic study and implications for the performance assessment of radwaste geological disposal. *Chem. Geol.*, **193**, 273-293.

Pescatore, C., Nachmilner, L., Martell, M., Mays, C. (2013): "Oversight of deep geological repository and the role of monitoring – Some preliminary findings within the RK&M Project of the NEA". Paper submitted for the MoDeRn International Workshop in Luxembourg, March 2013.

Petit, J.C. (1990): Migration of radionuclides in the geosphere – what can we learn from natural analogues? *Radiochim. Acta*, **51**, 181-188.

Polyak, V.J., and Asmerom, Y. (2001): Late Holocene climate and cultural changes in the southwestern United States. *Science*, **294**, 148-151.

Polyak, V.J., McIntosh, W.C., Guven, N., Provencio, P. (1998): Age and origin of the Carlsbad Cavern and related caves from $^{40}Ar/^{39}Ar$ of alunite. *Science*, **279**, 1919-1922.

Raney, R.G. (1988): Reported effects of selected earthquakes in the western North America intermountain region, 1852-1983, on underground workings and local and regional hydrology – a summary. *U.S. Nuclear Regulatory Commission Report* NNI-1988-3143, 114p.

Reverdatto, V. V. (1964): Manifestation of high temperature contact metamorphism of limestones in the basin of the Podkamennaya-Tunguzka river. *Dokl. Acad. Sci*., USSR **155**, 104–107.

Rizoulis, A., Milodowski, A.E., Morris, K., Lloyd, J.R. (2014): Bacterial diversity in the hyperalkaline Allas Springs (Cyprus), a natural analogue for cementitious radioactive waste repository. *Geomicrobio. J.*, DOI: 10.1080/01490451.2014.961107.

Robles-Camacho, J., and Armienta, M.A. (2000): Natural chromium contamination of groundwater at León Valley, México. *J. Geochem. Explor.*, **68**, 167-181.

Romberger, S.B. (1984): Transportation and deposition of uranium in hydrothermal systems at temperatures up to 300°C: Geological implications. *In*: (Vivo., B.D., Capaldi, F.I.G., Simpson, P.R., Eds.,) Uranium Geochemistry, Mineralogy, Geology, Exploration and Resources, London. **Institution of Mining and Metallurgy**, 12-17.

Savage, D. (2011): A review of analogues of alkaline alteration with regard to long-term barrier performance. *Mineral. Mag.*, **75**(4), 2401-2418.

Schindler, M., Fayek, M., Hawthorne, F.C. (2010): Uranium-rich opal from the Nopal I uranium deposit, Pena Blanca, Mexico: Evidence for the uptake and retardation of radionuclides. Geochimica et Cosmochimica Acta, 74, 187-202.

Schippers, A., Hallmann, R., Wentzien, S., and Sand, W. (1995): Microbial diversity in uranium mine waste heaps. *App. Environ. Microbio.*, **61**, 2930-2935.

Scholle, P.A., and Ulmer-Scholle, D.S. (1997): SEPM Photo CD-14, Environmental Sciences 5, SEPM, Tulsa, OK.

Shao, H.B., Kosakowski, G., Berner, U., Kulik, D.A., Mader, U., Kolditz, O. (2013) Reactive transport modeling of the clogging process at Maqarin analogue site. *Phys. Chem. Earth*. **64**, 21-31.

Smellie, J.A.T, and Karlsson, F. (1999): The use of natural analogues to assess radionuclide transport. *Engin. Geol.*, **52**, 193-220.

Sobolev, V. (1935): A rare type of contact metamorphism of limestones. *Azp. Vses. Min. Obshch*., **64**, 162-165.

Steefel, C.I., and Lichtner, P.C. (1998): Multicomponent reactive transport in discrete fractures II: Infiltration of hyperalkaline groundwater at Maqarin, Jordan, a natural analogue site. *Journal of Hydrology*, **209**, 200-224.

Stein, B.A., and Warrick, S.F. (1979): Granite Mountains resource survey – natural and cultural values of the Granite Mountains, Eastern Mojave Desert, California. Publication no. 1, University of California, Santa Cruz, Environmental Field Program, 363 p.

Stroes-Gascoyne, S., and West, J.M. (1996): An overview of microbial research related to high-level nuclear waste disposal with emphasis on the Canadian concept for the disposal of nuclear fuel waste. *Can. J. Microbio.*, **42**, 349-366.

Stroes-Gascoyne, S., and West, J.M. (1997): Microbial Studies in the Canadian nuclear fuel waste management program. *FEMS Microbio. Rev.*, **20**, 573-590.

Stroes-Gascoyne, S., Pedersen, K., Haveman, S.A., Dekeyser, K., Arlinger, J., Daumas, S., Ekendahl, S., Hallbeck, L., Hamon, C.J., Jahromi, N., Delaney, T-L. (1997): Occurrence and identification of microorganisms in compacted clay-based buffer material designed for use in a nuclear fuel waste disposal vault. *Can. J. Microbio.*, **43**, 1133-1146.

Stuckless, J.S. (2000): Archaeological analogues for assessing the long-term performance of a mined geologic repository for high-level radioactive waste. *U.S. Geological Survey* Open-File Report 2000-181, 27p.

Stuckless, J.S. (2002): Natural analogues—One way to help build public confidence in the predicted performance of a mined geologic repository for nuclear waste: Proceedings of the Waste Management 2002 Conference, Tucson, Arizona, February 24-28, 2002, v. 2002 symposium, 15p.

Stuckless, J.S. (2003): The use of qualitative analogues to help build confidence in predictive models for subsystems at Yucca Mountain, Nevada. *Geol. Soc. Am.* Abstracts with Programs, **35**, p.436.

Stuckless, J.S. (2006): Analogues as a means of facilitating communication with the public. *Geol. Soc. Am.* Abstracts with Programs, **38**, p.516.

Suksi, J., Ruskeeniemi, T., Saarinen, L. (1996): Selective extractions in uranium migration studies—findings from a natural analogue study at Palmottu, southern Finland. *J. Contam. Hydrol*., **21**, 47–58.

Toulhoat, P., Gallien, J.P., Louvat, D., Moulin, V., l'Henoret, P., Guérin, R., Ledoux, E., Gurban, I., Smellie, J.A.T., Winberg, A. (1996): Prelininary studies of groundwater flow and migration of uranium isotopes around the Oklo natural reactors (Gabon). *J. Contam. Hydrol.*, **21**, 3-17.

Taylor, J.H. (1935): A contact metamorphic zone from the Little Belt Mountains, Montana. *Am. Min.*, **20**, 120-128.

Techer, I., Khoury, H.N., Salameh, E., Rassineux, F., Claude, C., Clauer, N., Pagel, M., Lancelot, J., Hamelin, B., Jacquot, E. (2006): Propagation of high-alkaline fluids in an argillaceous formation: Case study of the Khushaym Matruk natural analogue (Central Jordan). *J. Geochem. Explor*., **90**, 53–67.

Techer, I., Bartier, D., Boulvais, Ph., Tinseau, E., Suchorski, K., Cabrera, J., Dauzères, A. (2012): Tracing interactions between natural argillites and hyper-alkaline fluids from engineered cement paste and concrete: Chemical and isotopic monitoring of a 15-years old deep-disposal analogue. *App. Geochem.*, **27**, 1384-1402.

Temple, A.K., and Heinrich, E.W. (1964): Spurrite from Northern Coahuila, Mexico. *Min. Mag*., **33**, 841-852.

Thomassin, J.H., and Rassineux, F. (1992): Ancient analogues of cement-based materials – stability of calcium silicate hydrates: *App. Geochem.*, Supplemental Issue, **1**, 137-142.

Tilley, C.E. (1929): On larnite (calcium orthosilicate, a new mineral) and its associated minerals from the limestone contact zone of Scawt Hill, County Antrim. *Min. Mag*., **22**, 77-86.

Tilley, C.E. (1933): Portlandite, a new mineral from Scawt Hill, County Anrim. *Min. Mag*., **23**, 419-420.

Tilley, C.E. (1942): Tricalcium silicate (rankinite), a new mineral from Scawt Hill. *Min. Mag.*, **26**, 190-196.

Tilley, C.E. (1947): The gabbro-limestone contact zone of Camas Mor, Muck, Inverness-shire. *Bull. Comm. Geol. Fin*., **20**, 97-105.

Tilley, C.E., and Harwood, H.F. (1931): The dolerite-chalk contact at Scawt Hill, County Antrim. *Min. Mag*., **22**, 439-468.

Toprak, V., Keller, J., Schumacher, R. (1994): Volcano-tectonic features of the Cappadocian volcanic province, Ankara, Turkey: 1994 International Volcanology Congress of the International Association of Volcanology and Chemistry of the Earth's Interior, Amsterdam, The Netherlands, Elsevier, 58p.

Tracy, R.J., and Frost, B.R. (1991): Phase equilibria and thermobarometry of calcareous, ultramafic and mafic rocks, and iron formations. *Rev. Min*., **26**, 207-289.

Tylecote, R.F. (1977): Durable materials for seawater – the archaeological evidence. The *Inter. J. Naut. Arch. Underw. Explor*., **6**, 269-283.

Tylecote, R.F. (1979): The effect of soil conditions on the long-term corrosion of buried tin-bronzes and copper. *J. Arch. Sci.*, **6**, 345-368.

U.S. Department of Energy, Earth Sciences Division: http://esd.lbl.gov/research/programs/new/research_areas/yucca_mountain/, accessed on 03/15/2015.

USGS, (2010): Analogues to features and processes of a high-level nuclear waste repository proposed for Yucca Mountain, Nevada. Professional Paper 1779, 194p.

Utada, M. (2003): Smectite-Zeolite Envelope Surrounding the Tsukiyoshi Uranium Deposit, Central Japan: A Natural Analogue Study. *Res. Geol.*, **53**, 293-304.

Valladas, H., Cachier, H., Maurice, P., Bernaldo de Quiros, F., Clottes, J., Cabrera Valdes, V., Uzquiano, P., Arnold, M. (1992): Direct radiocarbon dates for prehistoric paintings at the Altamira, El Castillo and Niaux caves. *Nature*, **357**, 68-70.

Villar, E., Bonet, A., Diaz-Caneja, B., Fernandez, P.L., Gutierrez, I., Quindos, L.S., Solana, J.R., Soto, J. (1985): Natural evolution of percolation water in Altamira cave. *Cave Sci., British Cave Res. Assoc.*, **12**, 21-24.

Watanabe, K. (1989): Archaeological evidence supports a preferred soil cap for LLRW disposal, in Proceedings of the 1989 Joint International Waste Management Conference, Kyoto, Japan, low and intermediate-level radiactive waste management. *Am. Soc. Mech. Engin.*, 567-571.

West, J., Alexander, W.R., Kaku, K., McKinley. I.G., Shimmura, A. (2002): Communication of nuclear power concepts to stakeholders - the use of nature's own laboratories. Proceedings for NUCEF 2001 - Scientific Bases for Criticality Safety, Separation Process and Waste Disposal. 31 Oct to 2 Nov 2001, Tokai, Japan. *Japan Atomic Energy Research Institute Report* JAERI Conf 2002-004., 47 – 54.

Wieland, E., and Spieler, P. (2001): Colloids in the mortar backfill of a cementitious repository for radioactive waste. *Waste Manag.*, **21**, 511-523.

Wikberg, P. (1987): The chemistry of deep groundwaters in crystalline rocks. Ph.D. Thesis: Department Inorganic Chemistry, KTH, Stockholm.

Wikipedia, Yucca Mountain: http://en.wikipedia.org/wiki/Yucca_Mountain_nuclear_waste_repository#/media/File:Yucca_Mountain_2.jpg, accessed on 03/14/2015.

Williamson, A.J., Morris,K., Law, G.T.W., Rizoulis,A., Charnock,J.M., Lloyd, J.R. (2014): Microbial Reduction of U(VI) under Alkaline Conditions: Implications for Radioactive Waste Geodisposal. *Environ. Sci. Tech.*, **48**, 13549-13556.

Wolf, J., Noseck, U., Meleshyn, A. (2013): Anwendbarkeit von Natürlichen Analoga für den Langzeitsicherheitsnachweis für ein Endlager im Salzgestein, *GRS* Report, forthcoming.

Woo, K.S., Hong, G.H., Choi, D.W., Jo, K.N., Bskaran, M., Lee, H.M. (2005): A reconnaissance on the use of the speleothems in Korean limestone caves to retrospective study on the regional climate change for the recent and geologic past. *Geosci. J.*, **9**, 243-247.

Wright, F.E. (1908): On the contact minerals from Velardena, Durango, Mexico (gehlenite, spurrite, hillebrandite). *Am. J. Sci*., **26**, p.156.

Wronkiewicz, D.J., Bates, J.K., Wolf, S.F., Buck, E.C. (1996): Ten-year results from unsaturated drip tests with UO_2 at 90°C: implications for the corrosion of spent nuclear fuel. *J. Nucl. Mater.*, **238**, 78-95.

Wyatt, M. (1953): The Camansury (Skye) gabbro-limestone contact. part II of Ph.D. thesis, Cambridge University.

Yamato, A., Masuda, S., Sakuma, H. (1992): The high-level radioactive waste management program in Japan, in Proceedings of the Third International High-Level Radioactive Waste Management Conference, Las Vegas, Nevada, April 12-16, 1992: *Am. Nucl. Soc.*, La Grange Park, Illinois, **1**, 41-48.

Yoshida, H., Yui, M., Shibutani, T. (1994): Flow-Path Structure in Relation to Nuclide Migration in Sedimentary Rocks. *J. Nucl. Sci. Tech.*, **31**, 803-812.

Zirngast, M. (1991): Die entwicklungsgeschichte des saltzstocks Gorleben- Ergebnisse einer strukturgeologischen bearbeitung. *Geol. Jb.*, **A132**, 3-31.