

# **GEOLOGIC DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTE IN SALT FORMATIONS**

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## **ABSTRACT**

Deep burial of high-level radioactive waste in salt formations is currently being studied as one potential solution for disposal of the nation's inventory of spent nuclear fuel and other heat-generating radioactive waste. This report provides a brief history and general overview of the concepts for geologic disposal of high-level waste and spent nuclear fuel in salt formations. Based on review of available literature, the aspects of potential salt disposal that provide the capability to isolate waste from the environment are highlighted and processes that may affect barrier capability are discussed.

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# 1 INTRODUCTION

The future direction of national policy for disposition of high-level radioactive waste (HLW), including spent nuclear fuel, is presently uncertain after the petition by the U.S. Department of Energy (DOE) to withdraw its license application for construction of a Yucca Mountain nuclear waste repository. The President of the United States chartered the Blue Ribbon Commission on America's Nuclear Future to review options for managing the back end of the nuclear fuel cycle, including alternatives for storage, processing, and disposal.

A key recommendation of the Blue Ribbon Commission's Draft Report (BRC, 2011) is to proceed with "prompt efforts to develop, as expeditiously as possible, one or more permanent deep geological facilities for the safe disposal of spent fuel and high-level nuclear waste." Geologic disposal in salt is one of several options for permanent disposal of HLW in which the Blue Ribbon Commission has expressed "substantial confidence." Additionally, the DOE National Nuclear Security Administration presently is funding an evaluation of salt disposal of heat-generating nuclear waste that includes international collaborations with the German salt disposal program. Hansen and Leigh (2011) outlined progress of this DOE-funded program in a recent report that summarizes the current state of knowledge about disposal systems in salt; highlights aspects of disposal in salt favorable to waste isolation; and discusses the potentially important features, events, and processes (FEPs) that should be considered for this disposal method.

Given the current uncertainty in the nation's HLW disposal program and the fact that federally funded agencies and committees are presently weighing the merits of salt disposal along with several other options, the purpose of this report is to provide an overview of the characteristics of salt formations in the context of geologic disposal of heat-generating waste and aspects of a salt repository system that may be important to safety and waste isolation.

Salt formations were among the first sites considered for possible HLW disposal because they exhibit natural properties favorable to long-term disposal of nuclear waste—low water content, tectonic stability, and high elasticity (National Academy of Sciences, 1957). Salt deposits were assumed to provide little or no mechanism and pathway for aqueous transport of radionuclides. The very existence of the rock salt was considered evidence of the separation of the salt formation from local circulating groundwater because otherwise the salt would have dissolved. Salt deposits also were assumed to be sufficiently elastic to absorb earthquake stresses and sufficiently plastic to be self-healing. Some of the favorable and unfavorable properties of a salt host rock identified in previous investigations are compared with properties of other types of geologic host media in Table 1-1. Whereas salt has several favorable properties, such as very low permeability and high visco-elastic creep, its high solubility and low sorption properties could affect its barrier capability in certain settings.

## 1.1 Previous Investigations of Salt for Geologic Disposal

In the 1970s and 1980s, several salt sites in the United States were investigated for potential HLW disposal, including bedded salt sites in Deaf Smith County, Texas; the Richton and Cypress Creek salt domes in Mississippi; and the Vacherie salt dome in Louisiana. Prior to drawing conclusions on site suitability, however, in-depth characterization of potential salt sites for HLW disposal was abandoned in 1987 when a Nuclear Waste Policy Act revision restricted consideration of a geologic repository to Yucca Mountain, Nevada.

<b>Property</b>	<b>Salt</b>	<b>Crystalline Rocks</b>	<b>Argillaceous Formations</b>
Thermal Conductivity	High	Medium	Low
Permeability	Very low	Very low (unfractured) to permeable (fractured)	Very low to low
Deformation Behavior†	Visco-plastic (Creep)	Brittle	Plastic to brittle
Strength	Medium	High	Low to medium
Dissolution Behavior†	High	Very low	Very low
Sorption Behavior†	Very low	Medium to high	Very high

\*German Federal Ministry of Economics and Technology. "Final Disposal of High-Level Radioactive Waste in Germany—The Gorleben Repository Project." <<http://www.bmwi.de/English/Redaktion/Pdf/final-disposal-of-high-level-radioactive-waste,property=pdf,bereich=bmwi,sprache=en,rwb=true.pdf>> (14 April 2011).  
†Does not include changes because of thermal perturbation.

Favorable

Average

Unfavorable

Salt deposits near Carlsbad, New Mexico, were investigated beginning around 1974 for disposal of transuranic waste generated by U.S. Department of Defense activities. These studies supported development and eventual certification of the Waste Isolation Pilot Plant (WIPP), which began accepting waste for disposal in 1999. To date, the WIPP site is the only deep geologic repository for radioactive waste in the United States. Thus, lessons learned from development of WIPP may be of great value toward evaluating the performance of salt for isolation of HLW. The transuranic waste and mixed wastes disposed at WIPP, however, do not generate significant amounts of heat compared to the current inventory of HLW. Therefore, in addition to the knowledge gained from operations at WIPP, the potential effects of heat-generating waste on the mechanical, chemical, and hydrologic properties of salt formations should be considered.

In Germany, there is operating experience from two deep geologic salt repositories for intermediate-level radioactive waste—both in former salt mines: the Schacht Asse II mine and the Bartensleben mine in Morsleben. A German Federal Ministry of Economics and Technology (2008) report drew the following conclusions on what it viewed as positive attributes of salt for disposal of HLW.

- Salt formations can serve as a geologic barrier that effectively isolates waste. Rock salt is known to react to mechanical loads with slow, flowing movements known as “creeping.” This causes cavities in the salt formation to be “self-sealed” or “healed.” Therefore, a natural geologic barrier function is guaranteed over very long periods in a salt repository system.
- Well over a century of industrial salt-mining experience and decades of intensive research both domestically and internationally have provided an extensive knowledge base regarding the properties of salt formations that are relevant for the final disposal of HLW.
- Mining experience has demonstrated that stable underground structures can be constructed in salt formations. The favorable mechanical properties associated with

rock salt allow the creation of cavities that do not require any special support structures and that can be maintained for decades.

- Salt formations exhibit low porosity and are impervious to gases and liquids, which are highly desirable attributes for any host rock.
- With a high level of specific thermal conductivity, rock salt makes for an effective host rock. HLW-generated heat [ $>110$  million  $m^3$ ] of petroleum in salt caverns at 4 sites near the Gulf of Mexico in Texas and Louisiana.

Posey and Kyle (1988) reviewed the various studies that had been conducted to evaluate the potential use of salt deposits for nuclear waste isolation. They stated that it was difficult to find documents that definitively either accept or reject salt domes as possible safe nuclear waste repositories. For example, they cited Knauth, et al. (1980), who found that brine seeps in the Weeks Island, Louisiana, salt dome originated as formation water from sources external to the dome. Thus, Posey and Kyle (1988) concluded that some salt domes are not as stable and dry as originally assumed, especially under mining conditions. On the other hand, Posey and Kyle (1988) also cited the work of Kreitler and Dutton (1983), who studied the Oakwood Dome in east Texas and the Gyp Hill Dome in south Texas. They found that the Oakwood anhydrite cap rock, which formed more than 100 million years ago, remains an effective hydrologic seal, whereas the Gyp Hill cap rock, which is still forming, is not an effective hydrologic seal and is allowing active dissolution of the salt dome. Thus, Kreitler and Dutton (1983) were more positive about using at least some salt domes for waste isolation if the salt dome has anhydrite cap rocks with the same characteristics as the Oakwood Dome cap rock.

At the Asse site in Germany, several sources of water inflow have been discovered that could ultimately necessitate movement of the waste to an alternative location at great expense (Sustainable Fuel Cycle Task Force, 2010). This experience at the Asse site underscores the need to fully understand the properties of the salt body and the potential for leakage or water penetration from or into adjacent water-bearing formations.

The experience gained using salt formations to contain petroleum reserves also is a relevant source of knowledge on the capability of salt to contain vast quantities of an environmentally hazardous liquid without significant leakage to the environment. The U.S. strategic petroleum reserve safely stores more than 700 million barrels [ $>110$  million  $m^3$ ] of petroleum in salt caverns at 4 sites near the Gulf of Mexico in Texas and Louisiana. Petroleum storage in salt requires continuous monitoring, and sites have been closed in the past when the integrity of salt formations became compromised. For example, Bayou Choctaw cavern 20 near Baton Rouge, Louisiana, was decommissioned in 2011 when geophysical surveys determined the stored petroleum to be within 18 m [60 ft] of the edge of the salt dome formation, compared to the federal requirement for a salt barrier of at least 91 m [300 ft]. In 1999, a petroleum storage facility in a former conventional room-and-pillar-type salt mine at Weeks Island, Louisiana, was decommissioned when a sinkhole brought fresh water into contact with the top of the formation, causing the potential for dissolution and erosion of the formation. These experiences with petroleum storage provide insights into understanding the barrier capability of salt formations and potential threats to formation integrity.

## 1.2 Barrier Functions for Waste Isolation in Salt

The previous investigations provide both positive and negative indicators of the suitability of salt to provide long-term containment of wastes, depending largely upon the characteristics of a particular site. Experience at any one site should not lead to conclusions regarding the suitability of all salt formations for disposal of radioactive wastes. For any potential candidate site, the barrier capability of a salt repository system is affected by following four site- and design-dependent characteristics that may enhance or degrade the barrier capability.

1. Permeability Barrier Effectiveness. The extremely low permeability of intact rock salt can prevent water from outside the formation from penetrating into it and fluids trapped inside the formation from migrating out. The effectiveness of the permeability barrier may be affected by the following site-specific or design-dependent processes or characteristics:
  - Deformation effects on salt formations
  - Near-field thermal and hydrological processes
  - Far-field hydrological processes
2. Brine/Water Mobility in the Repository Near Field. Existing brine pockets and fluid inclusions near the repository horizon could contact waste packages if they are present in sufficient quantity and mobilized by heat or hydraulic gradient through cracks or fissures in the repository disturbed zone. Brine/water mobility may be affected by the following site-specific or design-dependent processes or characteristics:
  - Fluid inclusions in salt formations
  - Near-field thermal and hydrological processes
  - Deformation effects on salt formations
3. Waste Package Integrity. Regardless of brine/water mobility within the formation or outside of the formation, waste can only be released if waste packages are breached. Waste package integrity may be affected by the following site-specific or design-dependent processes or characteristics:
  - Range of brine compositions, including effects of heat and radiation
  - Container materials and corrosion processes
  - Deformation effects on waste packages
4. Radionuclide Mobility. In the event of water contacting the waste and the existence of aqueous pathways through the formation, the mobility of radionuclides, including dissolution and transport processes, will determine the magnitude of release to the environment. Radionuclide mobility may be affected by the following site-specific or design-dependent processes or characteristics:
  - Range of brine compositions, including effects of heat and radiation
  - Container materials and corrosion processes
  - Near-field thermal and hydrological processes
  - Far-field hydrological processes
  - Radionuclide transport processes

The waste-isolation capability of salt formations is affected by a relatively small number of processes and characteristics, each of which has inherent uncertainties and complexities that will vary from one potential site to another. Several of these processes are common to more than one of the four main waste isolation characteristics. In a hot repository system, these processes may be interdependent, necessitating consideration of potential thermal-hydrological-mechanical-chemical (THMC) couplings. For example, THM coupling may affect the creep rate that, in turn, may cause changes in porosity and permeability in the near field; TC coupling may affect corrosion rates and the temperature-dependent solubility and sorption properties of the formation.

### 1.3 Report Organization and Scope

The barrier capability of the salt formation is impacted by its geological, geomechanical, hydrological, and geochemical properties as detailed in Chapters 2–4. The corrosion processes of potential waste container materials are evaluated in Chapter 5. As listed in Table 1-2, Chapters 2–5 of this report focus on discussion of these processes and their potential effects on waste isolation. Chapter 6 provides a summary, based on the preceding chapters, of the key process uncertainties for each of the four waste isolation characteristics enumerated previously.

This report does not consider potential disturbances due to disruptive events, such as seismic activity, or stylized scenarios, such as human intrusion, which could also affect waste isolation. In addition, this report does not include a detailed evaluation of FEPs related to waste disposal in salt formations (e.g., German and WIPP programs). These topics are recommended for future work. Technical challenges associated with retrieving waste in salt formations are expected to be different when compared to repositories in crystalline rocks or argillaceous formations. However, retrievability issues are not addressed further in this report, because these are dependent on policy decisions.

**Table 1-2. Organization of This Report**

<b>Chapter</b>	<b>Title</b>	<b>Topics covered</b>
2	Salt Geology and Geomechanics	<ul style="list-style-type: none"> <li>• Deformation effects on salt formation</li> <li>• Deformation effects on waste packages</li> </ul>
3	Hydrology and Radionuclide Transport Processes	<ul style="list-style-type: none"> <li>• Near-field thermal and hydrological processes</li> <li>• Far-field hydrological processes</li> <li>• Radionuclide transport processes</li> </ul>
4	Geochemical Processes	<ul style="list-style-type: none"> <li>• Fluid inclusions in salt formations</li> <li>• Range of brine compositions</li> <li>• Temperature effects on brine compositions</li> <li>• Radiation effect on brine chemistry</li> </ul>
5	Waste Package Corrosion	<ul style="list-style-type: none"> <li>• Container materials and corrosion processes</li> </ul>

## 2 SALT GEOLOGY AND GEOMECHANICS

The shape of a salt body and geometric relationships among its constituent layers or zones and adjacent geologic materials affect the spatial distribution of material properties (such as thermal and hydraulic conductivities) that may affect the barrier functions for waste isolation in salt. Salt formations consist primarily of the mineral halite and generally include one or more additional evaporite minerals (e.g., sylvite, gypsum, anhydrite, polyhalite, carnallite, trachyhydrite) along with carbonates (e.g., limestone, dolostone) and fine-grained siliciclastic (mudstone, siltstone, sandstone) sedimentary rocks (e.g., Blatt, et al., 1980; Hovorka, 2000; Liang, et al., 2007). Typically, salt is deposited as approximately horizontal layers, although irregularities in basin geometry can lead to spatially variable thickness. Some salt layers remain approximately planar after deposition (referred to as bedded salt deposits), whereas other salt layers develop into diapirs of various types (salt domes, salt pillars, salt walls). Examples of bedded salt deposits in the United States include Silurian-age deposits in the Michigan and Appalachian Basins (Michigan, West Virginia, Pennsylvania, New York, and Ohio), Devonian-age deposits in the Williston Basin (North Dakota and Montana), Permian-age deposits in the Delaware/Permian basins (Texas, New Mexico, and Oklahoma), and Cretaceous-age deposits in south-central Florida. Jurassic-age domal salt deposits are found in the onshore and offshore Gulf of Mexico region (Texas, Louisiana, Mississippi, Alabama, and Florida). Both bedded and domal Pennsylvanian-age salt deposits are found in the Paradox basin (Utah and Colorado).

The suitability of any particular salt formation for geologic disposal of high-level radioactive waste will depend largely on its physical and geomechanical characteristics. In particular, a suitable formation must be able to withstand the changes in stress state and resulting deformation that will occur in response to the excavated openings and the introduction of a significant heat source into the formation. In the following two sections of this chapter, the geomechanical characteristics of salt formations are discussed in terms of deformation effects on (i) the formation itself and its capability to maintain waste isolation and (ii) the waste packages and their response to mechanical forces within the formation.

### 2.1 Deformation Effects on Salt Formations

Geomechanical characteristics determine the potential formation of connected flow paths through deformation of the salt body. A salt body may deform in response to excavation of an underground opening; heat release from disposed nuclear waste; or other occurrences that tend to change the state of stress within the body, such as gas pressure buildup due to corrosion processes as detailed in Winterle, et al. (2011). The majority of constituents of a salt body, such as halite (rock salt), gypsum, and other evaporites, deform slowly through creep mechanisms when subjected to deviatoric stress (i.e., a stress state with different stress magnitudes in different component directions). The creep rate increases with the magnitude of deviatoric stress and with temperature. Because of the creep response of such constituents, underground openings in a salt formation can deform extensively over time, and in some cases, the opening can eventually close completely due to gradual deformation of the surrounding rock. For example, disposal openings at WIPP that were initially approximately 4 m [13 ft] high, 10 m [33 ft] wide, and 91 m [300 ft] long underwent a convergence of approximately 30 cm [1 ft] in the first year and 7.6–13 cm/yr [3–5 in/yr] thereafter (Vanderkraats and Patchet, 2009). The potential for such self-sealing is often cited as a desirable attribute of salt as a repository host rock.

The WIPP experience does not include thermal effects, because the disposed waste at WIPP does not produce sufficient decay heat to affect rock temperatures. For repository designs that include a significant decay heat from the disposed waste, deformation of the salt body will include the effects of increased creep rates due to higher temperature (e.g., Hansen and Carter, 1980; Carter and Hansen, 1983; Hansen, 2010) and the effects of stress change due to suppressed thermal expansion. Stress change due to thermal expansion results from the fact that heat from disposed nuclear waste will cause nonuniform temperature changes within the surrounding rock. Consequently, the rock tries to expand by different amounts in different directions at neighboring points. However, some of the potential expansion is suppressed due to neighborhood resistance, resulting in nonuniform increase in rock stress. As a result, the rock deforms to adjust to a new equilibrium state at a rate controlled by the prevailing creep rate.

The effects of salt deformation on the rock mass mechanical and hydrological properties depend on whether the prevailing stress states during the deformation favor dilation or compaction. Salt dilation (inelastic increase in volume) consists of growth and opening of microcracks and pores, results in an increase in porosity and permeability and a decrease in stiffness and load-bearing capacity, and is driven by stress states characterized by a high ratio of deviatoric stress to confining stress. In contrast, salt compaction (inelastic decrease in volume) consists of closure of microcracks and pores, results in a decrease in porosity and permeability and an increase in stiffness and load-bearing capacity, and is driven by stress states characterized by a low ratio of deviatoric stress to confining stress. Dilation is associated with damage, whereas compaction is associated with healing. Laboratory studies of the dilation and compaction tendencies of rock salt have led to defining a surface in stress space referred to as the dilatancy boundary to distinguish between stress states that favor dilation and stress states that favor compaction (e.g., Hunsche and Hampel, 1999; Hansen, 2003; Schulze, 2007; Alkan, et al., 2007). Stress states outside the dilatancy boundary favor damage, whereas stress states within the dilatancy boundary favor healing. Rock salt subjected to stress conditions within the dilatancy boundary may undergo mechanical compaction as well as actual healing of cracks and pores through geochemical mass transfer, such as solution and reprecipitation or crystallization (e.g., Hansen, 2003; Lux and Eberth, 2007; Schulze, 2007). Hansen (2003) reported laboratory tests on predamaged rock salt that showed significant recovery of tensile strength and ultrasonic wave velocity and decrease in permeability. Lux and Eberth (2007) also used ultrasonic (p-wave) velocity measurements to detect healing in laboratory specimens. The severity of damage may affect the healing rate or effectiveness. For example, Hansen (2003) noted that wave velocity recovery was slower in specimens that were more severely damaged. Also, the magnitude of confining stress may affect the healing rate, as laboratory data in Hansen (2003) indicated.

The tendency for dilatant deformation and, therefore, damage also can be affected by conditions that affect strain gradients within the rock. For example, rheological contrasts due to material nonhomogeneity or temperature gradients could increase strain gradients enough to induce dilation. Observations at the WIPP site (e.g., Park and Hansen, 2009) illustrate the deformation effects of material nonhomogeneity in the vicinity of contacts between rock salt and anhydrite layers, including cases of restrained salt deformation, fractured anhydrite that formed lenses within the more compliant salt, and fractured rock salt. To investigate the effects of rheological contrasts due to temperature gradients, Langer and Wallner (1998) performed numerical modeling of a hypothetical disposal design at the Gorleben salt dome. They concluded that areas near the top of the salt dome could fracture because of the modeled thermal loading, but the fractures would not propagate deep enough into the salt dome to be of concern.

Salt body deformations need to be evaluated to assess potential effects on fluid flow or the environment and loading of waste containers. Dilation could result in zones of microcracks or dilated pores that may coalesce to form faster flow paths. Such flow paths could enhance water penetration from, or water leakage into, adjacent geologic formations. Conversely, healing could result in gradual closure of existing cracks and pores, thereby disrupting existing flow paths formed through dilation. The volume of rock surrounding the repository openings that undergo dilation damage and the length of time that the damage persists depend on several factors, including heat load, formation physical and mechanical properties, size of the excavated openings, engineered support systems, and local stress fields. Therefore, to assess the waste isolation capability of a salt formation, the potential for damage and healing needs to be evaluated to determine whether the thickness of undamaged formation between the waste disposal zone and the accessible environment will be sufficient.

To evaluate the potential for mechanical processes to create enhanced flow paths, it is necessary to evaluate the potential coalescence of higher permeability zones that may result from deformation, the potential time of such occurrence, and the resulting permeability or porosity changes. Processes and their inherent uncertainties considered for such an evaluation include

- Steady-state creep rate for rock salt
- Mechanical properties of contiguous stiffer materials and contact interactions between such materials and rock salt
- Dilatancy boundary for rock salt
- Quantification of rock salt dilation
- Quantification of rock salt compaction and geochemical healing
- Change in rock salt stiffness due to damage, compaction, or geochemical healing
- Relationship among dilation, coalescence of damaged zones, and permeability
- Hysteresis in the dilation versus permeability relationships

Several of the uncertainties, particularly the quantification of dilation, compaction, and geochemical healing, could be considered as technical gaps because a considerable advancement of knowledge may be needed to resolve the remaining technical questions. For example, the unresolved questions regarding dilation are illustrated by a comparative study of several rock salt constitutive models by Hou, et al. (2007). An analysis of predictions calculated using the models for two benchmark problems showed that while the model predictions are reasonably consistent when mechanical response is dominantly nondilatant, the predictions for conditions of dilatant deformation are more divergent as the amount of dilation increases (Hou, et al., 2007). Such divergence could have considerable implications for the assessment of potential effects of deformation on fluid flow: as laboratory testing by Peach and Spiers (1996) indicated, a small amount of dilation (e.g., <0.2 percent) could cause a large (e.g., five orders of magnitude) increase in permeability because of the effects of dilation on the connectivity of open cracks (e.g., Alkan, 2009).

## 2.2 Deformation Effects on Waste Packages

Deformation of salt could also result in external pressure on waste containers, if the host rock converges into the openings and contacts the containers, as suggested by essentially all existing conceptual descriptions for geologic disposal systems in salt formations. The amount of pressure exerted on a container will be determined by the depth of overburden combined with additional forces resulting from local heat effects and regional stresses. The effect of the external pressure on the waste packages will depend largely on the waste package design and whether it is intended to withstand the *in-situ* formation pressures. In the case of the WIPP site, for example, most of the transuranic waste disposed there is packaged in thin steel drums that are intended to eventually be crushed by the converging salt formation; therefore, no barrier function is intended or credited for waste package protection at the WIPP facility. For high-level radioactive waste, it is more likely that waste packages would be designed to provide added waste isolation capability beyond that of the salt formation alone—at least for some initial period, until the more highly radioactive isotopes have decayed significantly. Some additional discussion of waste package materials and degradation is provided in Chapter 5 of this report.

Because heat can greatly increase the deformability of the salt directly surrounding the package, another factor that must be evaluated is the potential for the heavy containers to sink into the floor rock. This potential sinking could result in progressive mobilization of heated salt under the weight of the container, where forces would tend to mobilize the salt upward around the package, thereby increasing downward pressure on the container. As documented in Organisation for Economic Co-operation and Development/Nuclear Energy Agency (2000), the WIPP project considered potential effects of buoyancy forces on waste containers due to thermally induced density changes in the salt, but these were screened out because of the low heat-generating capacity of the containers. However, the effects of such density changes could be more significant for waste containers with greater heat-generating capacity. The process could result in the container being gradually submerged in the salt during the period of high temperatures, potentially exiting from the salt formation to an underlying nonsalt geologic material. If appropriately considered during repository design, this potential sinking problem could be controlled by ensuring sufficient formation thickness, managing heat loads, and limiting the bulk density of the waste packages.

For any particular site or design, salt deformation needs to be evaluated to assess potential interaction between a waste container and the surrounding rock. The processes and their inherent uncertainties to be considered include

- Steady-state creep rate for rock salt
- Mechanical properties of contiguous stiffer materials and contact interactions between such materials and rock salt
- Backfill compaction and geochemical healing and their effects on backfill stiffness
- The effects of heat on salt mechanical properties and deformation rates

## **3 THERMOHYDROLOGICAL AND RADIONUCLIDE TRANSPORT PROCESSES**

The primary objective of a salt repository system is to isolate waste from circulating groundwater in any adjacent layers with non-negligible groundwater flow, especially any aquifers above the salt deposit. Groundwater flow (both local and regional) adjacent to a salt formation, as well as the potential for water to enter into a salt formation, is influenced by factors such as climate, recharge, and geologic-mechanical-chemical conditions that extend well beyond the boundaries of the repository. Both ambient and thermally induced mechanical (e.g., fracturing) and geochemical (e.g., dissolution) effects could affect hydrological properties and in turn influence groundwater flow and radionuclide transport. Therefore, geomechanical (Chapter 2), geochemical (Chapter 4), and thermohydrological properties (Section 3.1) of the salt formation and the neighboring aquifers (overlying and underlying units) should be characterized appropriately and the coupled effects induced by heat load must be considered.

This chapter discusses potential factors that can affect hydrological processes and, hence, barrier performance, for a salt repository. The hydrological processes that may affect repository performance can be divided into near field and far field, which are discussed separately in the following two sections. Processes affecting transport of radionuclides in the near field and far field are discussed in the third section.

### **3.1 Near-Field Thermohydrological Processes**

In this section, the near field is described as the volume of rock influenced by excavation- or heat-induced changes in stress and hydrologic conditions. In an ideal, stable salt formation, there is expected to be essentially no flow and radionuclide migration would be limited to diffusion through very limited pore space (International Atomic Energy Agency, 2003). Near-field perturbation of the salt system from excavation and emplacement of heat-generating waste, however, may lead to geomechanical and geochemical changes that could affect hydrologic properties and enhance brine migration. Water is found within salt formations as intergranular and intragranular inclusions as well as in larger volumes in the form of brine-filled pockets. Such water can migrate within the salt in response to changing conditions. Observations of brine migration in mined salt formations are not uncommon and have been reported in excavations at WIPP and the Asse and Morsleben Mines in Germany (Larson, 2000; Sustainable Fuel Cycle Task Force, 2010). Geckeis and Klenze (2009, Section 5.2) reported that intrusion of 10 m<sup>3</sup>/d [350 ft<sup>3</sup>/d] of NaCl-rich brine into the Asse mine had been observed for several years. No information on the processes causing the intrusion of brine into the Asse mine or Morsleben sites has been found in the literature beyond the mention of the brine intrusion volumetric rate and duration. In excavations at WIPP, weeps on walls, puddles on floors, and brine infilling boreholes were analyzed and found to be similar to intergranular water associated with anhydritic and argillaceous material (Stein and Krumhansl, 1986; Larson, 2000).

Perturbations in the near field link the coupled processes of hydrology, thermohydrology, geomechanics, and geochemistry. Hansen and Leigh (2011) and Tsang, et al. (2005) describe coupled processes that may occur throughout the time evolution of the damage zone around excavations. The near-field disturbed zone plays a role in all phases of repository evolution. Wieczorek, et al. (2010) found the following potentially important issues unresolved:

- Evaluation of shaft-sealing concepts with respect to safety assessment regarding the repository design and the closure phase

- Backfill behavior during the closure and postclosure phases and in altered conditions
- Gas transport and storage during the postclosure phase
- Excavation damage zone evolution and recompaction during all phases until disturbed (altered) conditions have dissipated
- Brine migration in disturbed (altered) conditions

Wallner, et al. (2007) highlighted the uncertainty surrounding thermal-hydrological-mechanical evolution of the excavation damage zone and backfill. The primary issues they cited are the extent of brine migration toward waste packages, the spatial extent of the near field, and the length of time for the system (both the disturbed zone and backfilled seals) to return to natural hydrologic and geomechanical conditions.

Field and laboratory experiments at various temperatures ranging up to 250 °C [482 °F] have been performed (Table 3-1). Thermal gradients induced by waste heat can cause various types of geochemical perturbations. For example, solubility will increase slightly on the heated side of a brine pocket, resulting in dissolution on the hot side and precipitation on the cool side of the brine pocket that causes the brine to gradually migrate toward the heat source. Hansen and Leigh (2011) cite several examples of *in-situ* experiments in which such brine migration toward the heat source was observed. Mass transfer of water in the vapor phase can result from evaporation in hot regions and advection to cooler regions where condensation of the water vapor to liquid can occur. This water vapor could condense on waste containers and in the host salt formation in cooler areas of the repository. This could (i) affect corrosion rates and gas generation associated with the corrosion process and (ii) potentially dissolve the host rock because of the relatively lower dissolved salt concentration in the condensate. Thermal perturbation also may have the favorable effects of speeding the healing/sealing rate of the damage zone and crushed salt backfill.

Excavation results in a damage zone of microfractures within a few centimeters [1 cm = 0.39 in] of the drift wall that could provide a pathway for fluid migration. The presence of an excavated opening in a previously stable formation also disturbs the local stress fields. As discussed in Chapter 2, dilation tendency is most intense within a zone around the openings and dilation (volume increase) can result in enhanced permeability and the coalescence of fractures in the excavation damaged zone. Backfilling after emplacement of waste can help return the system to natural conditions faster than if no backfill is used by reducing the open volume that must be filled by rock wall deformation. The rate at which the salt system returns to natural conditions is uncertain and has been the focus of many laboratory and field experiments (e.g., Bechthold and Hansen, 2003). If crushed salt is used as a backfill, the hydrological regime in the near field will be influenced by the range of porosity and hydraulic conductivity of the backfill, which are in turn dependent on the particle size distribution of the crushed salt. The added effects of heating and subsequent cooling result in stress changes that may lead to new fracturing, or the opening of interconnecting of existing fractures, depending on formation properties.

The near-field disturbed zone is conceptualized generally to be limited in extent and to eventually heal to a state approaching *in-situ* permeability as the creep processes cause openings and fractures to gradually close. The period in which the near field remains in a disturbed condition is highly uncertain, depending on formation properties and heat load, but

<b>Table 3-1. Thermal Tests in Salt</b>			
<b>Site</b>	<b>Name of Test</b>	<b>Description and Focus</b>	<b>Maximum Temperature</b>
Waste Isolation Pilot Plant (WIPP)*†	Defense High-Level Waste Mockup	Vertical boreholes in floor; evaluated mechanical response to thermal load	—
WIPP*†	Defense High-Level Waste Overtest	Vertical boreholes with heaters at two different supplied power levels; evaluated of crushed backfill and collection of brine for periods up to 600 days	250 °C [482 °F]
WIPP*†	Heated Axisymmetric Pillar Test	Evaluated scale effects for thermal-mechanical processes	70 °C [158 °F]
WIPP‡	Crushed Rock Reconsolidation Test	Evaluated extent and rate of reconsolidation of crushed salt backfill in the laboratory	250 °C [482 °F]
Louisiana*	Avery Island Salt Mine	Heated borehole test with supporting laboratory analyses; evaluated migration of brine inclusions; collected moisture released during test; concluded water released during cooling stage	51 °C [123 °F]
Asse Mine§	Thermal Simulation of Drift Emplacement (TDSE)	Two heated drifts, posttest excavation of one drift; evaluated healing rate and extent of damage zone and backfill	170 to 210 °C [338 to 410 °F]
Asse Mine	Development of Borehole Seals for Radioactive Waste (DEBORA)	Deep borehole concept, performance of crushed salt; evaluated rate of compaction, change in permeability, and extent of porosity reduction	140 °C [284 °F]
Russian Lab Test¶	—	Two intermediate-scale laboratory tests designed to study brine and vapor distribution and migration around heater and in backfill	200 °C [392 °F]

\*Hansen, F.D. and C.D. Leigh. "Salt Disposal of Heat-Generating Nuclear Waste." SAND2011-0161. Albuquerque, New Mexico: Sandia National Laboratories. 2011.

†Matthews, M.L., and L.G. Eriksson. "The Waste Isolation Pilot Plant—An International Center of Excellence for Training in and Demonstration of Waste Disposal Technologies." Proceedings From the WM'03 Conference, February 23–27, 2003, Tucson, Arizona. 2003.

‡Clayton, D.J., M.Y. Lee, D.J. Holcomb, and D.R. Bronkowski. "Crushed Salt Reconsolidation at Elevated Temperatures." ARMA 10-236. Proceedings from the 44<sup>th</sup> U.S. Rock Mechanics Symposium, June 27–30, 2010, Salt Lake City, Utah. 2010.

§Rothfuchs, T., W. Gollingerfehr, and W. Bechthold. "Lessons Learned From Large-Scale Experiments at the Asse Mine/Germany." Paper 6-7. Proceedings From Invited Lecture EURADWATE '04, Luxembourg, March 29–31, 2004.

||Bechthold, W. and F. Hansen, eds. "Final Report—Backfilling and Sealing of Underground Repositories for Radioactive Waste in Salt, Phase II (BAMBUS II)." EUR 20621. European Commission, Nuclear Science and Technology. 2003.

¶Schneider, L.R. "*In-Situ* Experiments, HLW-Investigations: Russian Salt Experiments." Presentation at U.S.-German Workshop, Jackson, Mississippi, May 25–28, 2010.

could persist for several thousand years. During the operational period of a repository, any accessible brine will tend to migrate down the stress gradient and evaporate into the ventilation air. After closure, as salt creeps, the stress gradient decreases, fractures heal, and crushed salt backfill (if present) reconsolidates. This reconsolidation is expected to significantly limit brine flow to the waste disposal areas once the healing is complete. However, there is uncertainty regarding the heterogeneity of the creep process and this, in turn, could result in some repository locations that could experience brine flow for extended periods. This potential for brine flow should be evaluated in detail in terms of its effect on flow and transport of radionuclides.

Excavation and subsequent salt creep result in dilation (volume expansion) of host rock that eventually leads to a decrease in fluid pressure (Hansen and Leigh, 2011). If the pores dilate more than the brine can expand, partially saturated conditions may result. Dissolved gas may come out of solution as the pore pressure decreases, resulting in two-phase conditions. The effect of dilation on permeability can be evaluated using the percolation theory proposed by Peach (1991). Under undisturbed conditions, the low porosity of rock salt may be too poorly connected to reach the minimum pore connectivity at which flow can occur (the percolation threshold). With relatively small amounts of dilation ( $\geq 0.05$  percent of volume), however, Peach (1991) found that sufficient grain-boundary microfracturing occurs to create an interconnected pore network beyond the percolation threshold. From the percolation threshold, permeability increases rapidly with increasing dilation until the pore network is fully interconnected, which occurs at some dilation less than 1 percent. After the pore network is fully interconnected, increased dilation has little effect on permeability. The dilatancy of halite is enhanced by the presence of impurities (e.g., anhydrite, clay), so impure halite will dilate more and thereby increase in permeability to a greater distance from an excavation than will pure halite. Studies at the WIPP site indicate that permeability is extremely sensitive to small, dilatant volumetric strains. Permeability within the disturbed zone could increase by three to seven orders of magnitude in response to the dilation (Hansen, 2003).

The permeability created (or enhanced) by excavation effects is anisotropic and has a strong directional dependency (Beauheim and Roberts, 2002). Fractures in salt develop preferential orientation with respect to the stress state, parallel to the drift and perpendicular to the minimum principal stress (Hansen and Leigh, 2011). Permeability along the fractures parallel to the drift would be much higher than permeability perpendicular to the drift. The anisotropic nature of fractures could complicate development of a damage/permeability relationship for field applications.

### **3.2 Far-Field Hydrological Processes**

For purposes of this report, the far field is defined to include portions of the salt formation outside the near-field disturbed zone and any other overlying or underlying formations that may lie between the repository and the accessible environment or compliance boundary.

Based on the field and laboratory studies at WIPP and in Germany, hydrological processes in the far field are not likely to directly affect the emplaced waste (Beauheim and Roberts, 2002; Noseck, et al., 2009). That is, when a salt repository performs as conceptualized, the waste should remain isolated from water flow and transport pathways in the far field by the undisturbed portions of the salt formation above and below the emplaced waste. The undisturbed portions of the salt formation surrounding the disposal zone are arguably the principal barrier to waste migration in a salt repository system. Potential threats to this isolation could occur due to flaws in design implementation (e.g., poorly sealed shaft); extension of the disturbed zone

(e.g., dilation damaged) to overlying or underlying permeable layers; or erosion or dissolution of the salt formation (e.g., due to climate-induced changes in hydrologic regime at the surface).

In undisturbed salt formations, porosity of both halite and anhydrite layers is typically measured to be very low (Beauheim and Roberts, 2002; Liang, et al., 2006, 2007). In bedded salts, anhydrites and clay-rich layers have the relatively highest permeabilities and pure halites have the lowest permeabilities. Anhydrite permeability derives from naturally occurring bedding-plane fractures (Borns, 1985). The extremely low permeability in halites impedes vertical flow, and thus, to the extent that any flow occurs, it is more likely to occur parallel to the bedding along layer interfaces or zones of high impurities. Within an individual bed, flow may be channelized into relatively permeable pathways created either by natural heterogeneity or by syndepositional dissolution troughs filled with pure halite (Powers and Hassinger, 1985) cutting across the bed that limit flow in a certain direction.

Beauheim and Roberts (2002) estimated groundwater travel times in halite and brine at the WIPP site. Assuming a halite permeability of  $10^{-22}$ – $10^{-21}$  m<sup>2</sup> [ $10^{-10}$ – $10^{-9}$  darcy] {hydraulic conductivity of  $10^{-15}$ – $10^{-14}$  m/s [ $3 \times 10^{-15}$ – $3 \times 10^{-14}$  ft/s]}, a hydraulic gradient of 0.01, and a porosity of 0.01, they estimated that brine would take between 3 million and 30 million years to flow 1 m [3 ft]. In anhydrite, where the permeability might be 100 times higher, they calculated that brine would still take 30,000 to 300,000 years to flow 1 m [3 ft].

However, there is conceptual uncertainty in the calculation of groundwater travel times based on Darcy's Law, which assumes that groundwater flow is driven by pressure gradients for most porous media. Some researchers claim that Darcy flow has never actually been demonstrated in low-permeability formations, such as the Salado, under natural pressure gradients (Beauheim and Roberts, 2002). Other researchers (e.g., Swartzendruber, 1962; Pascal, 1981; Remson and Gorelick, 1982) have suggested that low permeability media may have threshold gradients below which no flow can occur, perhaps related to capillarity or surface-tension effects in very small pores. Gas-threshold (air-entry) pressures of the evaporites are high, probably exceeding the hydraulic-fracturing pressures, because of the very low permeabilities. To whatever extent regional flow is occurring under the naturally occurring pressure gradients, it is expected to be toward areas of lower overburden pressure.

Owing to their low permeability, most salt domes are located below the water table as percolating groundwater tends to pool on top of such relatively impermeable formations. As such, any overlying formations with sufficient permeability could be potential sources for agricultural or drinking water. Factors that may affect hydrological processes above the salt formation include climate, recharge, and saline and freshwater intrusion. Changes in the surface environment driven by climate change influence the recharge to the groundwater. The potential effects of surface-water bodies, such as rivers, lakes, ponds, or streams, on local groundwater recharge should be evaluated. Climate-related factors that might affect groundwater flow, such as precipitation, temperature, and evapotranspiration, also should be evaluated for the site. Changes to these climate-related factors for a range of different future states in which the climate may be wetter could result in water table rise and increased groundwater velocity.

Above the water table, flow conditions in the unsaturated zone should be evaluated for estimates of the recharge. Changes in climate or recharge could result in freshwater intrusion into the host rock and result in dissolution processes (detailed next) or decrease the fluid density in the host rock. Direct contact with groundwater could lead to formation of a zone of highly concentrated brines at the top of the salt formation (Klinge, et al., 2002). Depending

on the concentration gradients and general groundwater regime, the movement of concentrated brines could be very important in determining the radionuclide migration in the overlying units. In the absence of a driving hydraulic gradient, however, brine zones atop salt formations will have a higher density than any overlying fresh water and tend to remain stable in place. Additionally, the presence of confining units, such as clay layers above the salt, also hinders the formation of flow cells and prevents the intrusion of brines into the overlying formations. As mentioned previously, the fact that soluble salt formations persist for millions of years is itself evidence of limited interaction with circulating fresh water sources.

In places where uplift, erosion, or changes in groundwater flow patterns have brought salt formation into contact with fresh water circulation, dissolution of the salt can occur. It is therefore important to evaluate the potential for shallow, lateral, or deep dissolution when selecting a potential salt repository location. Shallow dissolution involves percolation of groundwater and mineral dissolution in a rock formation that overlies the host rock formation. Lateral dissolution refers to dissolution of the top of the host rock formation by percolating groundwater. Both shallow and lateral dissolution could result in collapse of the overlying units (Organisation for Economic Co-operation and Development/Nuclear Energy Agency, 2000). Extensive dissolution may create large cavities (karst) and result in the collapse or subsidence of overlying units.

Deep dissolution refers to the dissolution of salt or other evaporite minerals in a formation at depth. Deep dissolution is distinguished from shallow and lateral dissolution not only by depth, but also by the origin of the water, which in this case is groundwater from deep water-bearing zones. Deep dissolution can create cavities in the formation at depth, leading to collapse of overlying beds and to the formation of collapsed breccias (if the overlying rocks are brittle) or to deformation (if the overlying rocks are ductile). If dissolution is extensive, breccia pipes or solution chimneys may form above the cavity. Deep dissolution is of concern because it could accelerate contaminant transport by creating vertical flow paths. If dissolution occurred within or beneath the emplaced radioactive waste, there also could be increased circulation of groundwater through the waste itself. Evaluating the potential for such occurrence requires a thorough understanding of site characteristics.

If the local hydrologic regime is such that the pressure and gravity-driven components are low, then the groundwater density gradients could be the factors that determine the direction of groundwater flow. Variability in the brine composition at the site should be evaluated and incorporated in groundwater flow models if deemed important to density-driven flow.

### **3.3 Radionuclide Transport**

If radionuclides are released from a deep geologic repository, water provides the most likely pathway for radionuclide transport, but the low water content of rock salt could limit the transport of dissolved radionuclides. Evaporation of brines due to elevated temperatures during the thermal period, and the self-sealing capacity of the salt in general, could also limit the possibility for radionuclide releases and transport away from the repository. As Hansen and Leigh (2011) noted, investigations of the potential for radionuclide transport in a salt repository assume that there will be brine available to dissolve and transport the radionuclides. In practice, however, transport depends on at least two factors: brine sources with sufficient volume and the existence of a pressure gradient capable of moving the soluble radionuclides through the rock to the biosphere.

At the very low permeabilities associated with undisturbed salt formations, there is essentially no advective movement of water, so radionuclide transport away from the repository is limited to diffusion in whatever brine may be present in interconnected pathways (e.g., along grain boundaries) in the salt. Diffusion is the process whereby the constituents (e.g., ions or molecules) of a medium move under the influence of their kinetic activity in the direction of their concentration gradient (i.e., from high to low concentrations), with diffusion ceasing when the concentration gradient becomes nonexistent (Freeze and Cherry, 1979). Diffusion of dissolved constituents (e.g., ions or molecules) in water is influenced by temperature and by properties of the dissolved constituent, such as ionic charge and radius, which are expressed by a free-water diffusion coefficient that can be measured empirically or estimated from the atomic properties of the dissolved constituent.

Compared to diffusion of solutes in open water, diffusion through a solid medium is further slowed by (i) physical characteristics of the solid phase, such as its porosity and permeability; (ii) constrictivity, which results from the size of pore throats (i.e., the connections between adjacent pores) and how small pores are distributed in the medium (Katsube, et al., 1986); and (iii) tortuosity, which is the length of the transport path traced by a solute molecule as it winds through the interconnected pore space in the solid, as compared to the direct transport pathway that would be available to the solute in an unrestricted aqueous medium (Saripalli, et al., 2002). Tortuosity and constrictivity are difficult to measure directly, so the physical characteristics of a solid medium are typically represented by an empirically measured effective diffusion coefficient (Freeze and Cherry, 1979). Reimus, et al. (2007) used a quantitative relationship between porosity, permeability, and tortuosity to estimate the effective diffusion coefficients for solutes in welded volcanic tuffs, but the usefulness of the relationship diminished in rocks that had low porosity or low permeability. In an *in-situ* diffusion experiment that more closely resembles transport conditions in salt formations, Vilks, et al. (2003) measured field-scale diffusion rates in a deep {450 m [1,500 ft]}, highly stressed, sparsely fractured granite with low porosity (less than 0.01) and low permeability { $10^{-21}$  m<sup>2</sup> [ $10^{-9}$  darcy]}. The experiment measured effective diffusion coefficients that varied from  $2.1 \times 10^{-14}$  to  $1.9 \times 10^{-13}$  m<sup>2</sup>/s [ $6.9 \times 10^{-13}$  to  $6.2 \times 10^{-13}$  ft/s]. The highest diffusion rates were those within a centimeter [0.39 in] of the borehole openings. Diffusivity decreased markedly beyond that distance, an effect that Vilks, et al. (2003) attributed to local stress redistribution and creation of a zone of increased constrictivity close to the borehole wall. Similarly, Hansen and Leigh (2011) identified that the disturbed rock zone adjacent to excavations in salt formations would have increased permeability and interconnected porosity, potentially leading to brine flow into the excavation and higher transport rates away from the excavation for a short, reversible time period.

For some solutes, diffusive transport may also be retarded by adsorption of the solute onto solid surfaces. Generally speaking, the sorption capacity of salt minerals is low, but sorption could be more important if impurities or interbedded layers, such as clays, are present in the salt formations. In modeling the effects of sorption, the high ionic strength (total dissolved solids) in brine solutions complicates the use of sorption coefficients ( $K_d$ s) for radionuclides because many solutes are competing for the sorption sites on a solid at the same time (National Research Council, 1996).

Although the transport of radionuclides from a repository in a gas phase is expected to be less significant than their transport in water (brine), researchers have identified pressure buildup of hydrogen gas due to anaerobic corrosion of steel container materials as a potential concern. In the near field, pressure buildup could inhibit rock convergence and consolidation of crushed rock backfill, depending on the associated volume of hydrogen and gas generation rates (Hansen and Leigh, 2011). However, a field-based experiment in Germany measured

significant gas diffusion rates in salt, even in areas beyond the excavation damage zone. The experiments also found that gas diffusion coefficients depended on the molecular weight of the gas components and that the low water content of the salt prevented diffusion of gas dissolved in water (Jockwer and Wieczorek, 2008).

Although diffusion is expected to significantly limit the rate of radionuclide transport in salt formations, whenever a radionuclide reaches the formation boundary it will encounter potentially different transport conditions in a different rock type. Such factors are site specific and fall outside the scope of this report, but they would be considerations in evaluating the overall performance of the natural barriers in a deep geologic repository in a salt formation. In addition, layers of other rock types that are interbedded with a salt formation may locally provide pathways with different transport conditions and properties. Diffusion lengths to such pathways could be relatively short, compared to the thickness of the salt formation as a whole. This situation is conceptually analogous to a competent, sparsely fractured granite, in which radionuclide transport through the rock matrix is slow, due to low porosity and low permeability, but may increase significantly after the radionuclide enters a fracture pathway that is more permeable.

The other factors affecting radionuclide transport depend on perturbations to the initial environment, such as scenarios in which new pathways for flow and transport develop from natural dissolution of the salt or from human intrusion. In the performance assessment analysis for WIPP, the primary contributor to any possible release of radionuclides was human intrusion, so human-initiated features, events, and processes were studied extensively (Hansen and Leigh, 2011). In the WIPP case, the intrusion event was envisioned as drilling through a repository and creating a vertical flow pathway for water from depth and advective transport of radionuclides from the disrupted wastes. However, human intrusion scenarios and their effects would be site specific and are beyond the scope of this report.

## 4 GEOCHEMICAL PROCESSES

A potentially important factor affecting the performance of HLW repositories is the chemistry of waters in the near-field environment. Water chemistry will affect the corrosion of waste container materials, the dissolution of waste forms, and the release and transport of radionuclides from the near-field environment. Given the uncertainty in the degree to which salt formations are dry or hydrologically isolated, information is needed on the potential range in the chemistry of brines that may be present in salt formations. This information will allow an evaluation of the potential effect of these brines on waste form and waste container performance and on near-field transport. Published information is available on the chemistry of brines from salt formations previously considered for disposal of U.S. HLW. In the 1980s, DOE considered seven sites for the permanent disposal of HLW in salt formations (Jain, 1986):

- Richton Dome Site (Perry County, Mississippi)
- Vacherie Dome Site (Webster and Bienville Parishes, Louisiana)
- Cypress Creek Dome Site (Perry County, Mississippi)
- Lavender Canyon Site, Paradox Basin (San Juan County, Utah)
- Davis Canyon Site, Paradox Basin (San Juan County, Utah)
- Deaf Smith County Site, Palo Duro Basin (Permian, Texas)
- Swisher County Site, Palo Duro Basin (Permian, Texas)

These sites can be categorized into two groups depending on how the salt was formed or subsequently deformed: salt domes (the first three sites) and bedded salts (the last four sites). On average, bedded salts are believed to contain a volume of brine inclusions that is 10 times greater than dome salts. Site-specific differences also are present within each group. For example, the salt minerals carnallite ( $\text{KCl}\cdot\text{MgCl}_2\cdot 6\text{H}_2\text{O}$ ) and kieserite ( $\text{MgSO}_4\cdot\text{H}_2\text{O}$ ) are present in significant amounts in the Paradox Basin salt, but not in the Palo Duro Basin salt; both are bedded salts (Jain, 1986).

Another factor relevant to the assumed dry condition of salt formations is the presence of intragranular water in salt minerals, typically in the form of fluid inclusions or as waters of hydration in hydrous minerals. These waters may get mobilized as liquid or steam in response to thermal gradients HLW generates and accumulate at the hot waste container surface. This process, in addition to water intrusion from overlying groundwater aquifers, may cause corrosive brines to contact the HLW waste containers and waste forms.

### 4.1 Fluid Inclusions in Salt Minerals

Salt formations typically contain inclusions or cavities of brine solution. When seawater evaporates causing salts to precipitate, small volumes of water commonly are trapped in the salt. Some fluid inclusions have maximum dimensions measured in millimeters, but maximum dimensions are typically on the order of hundreds of microns and most are much smaller (Roedder, 1984a). Such fluid inclusions can form within individual salt crystals or along boundaries between salt crystals. Fluids within inclusions may represent original seawater from which the salt precipitated (Lowenstein, et al., 2001; Horita, et al., 1991, 2002), water introduced later into the formation, or water modified after deposition of the salt (Lowenstein, et al., 2005). These differences can be used to infer whether the salt formation has been affected by postdepositional fluids or processes (Organisation for Economic Co-operation and Development/Nuclear Energy Agency, 2008). Fluid inclusions can provide important information about the history of the salt deposit, including indications of past water

movement into the deposit, which may indicate the potential for future water introduction to the salt formation (Krupp, 2005).

Salt recrystallizes readily under the influence of a variety of low temperature diagenetic processes. As a result, most crystals now present in salt formations are not those formed by the original precipitation from seawater. As recrystallization occurs, fluids included within the crystals tend to coalesce into fewer, larger inclusions. Depending on the timing and circumstances of the recrystallization, the composition of the fluids may be similar to or different from the original fluids (Roedder, 1984a). Identification of brine pockets or zones with large quantities of fluid is difficult and contributes to the uncertainty for locating emplacement areas. Because of salt flow during formation of a dome and consequent squeezing out of included fluids, largely by migration along grain boundaries (Baes, et al., 1983), dome salts tend to contain less water as fluid inclusions (<0.1 wt%) than bedded salts (>1 wt%) (Roedder and Bassett, 1981).

Compositions of the fluids in inclusions vary from essentially pure NaCl solutions to solutions dominated by variable amounts of CaCl<sub>2</sub> and MgCl<sub>2</sub>, along with many other components in low amounts. Compositions of the salts reflect the degree of evaporation of the seawater at the time of trapping, introduction of water resulting from diagenetic effects (e.g., dehydration of gypsum), or addition of water from sources outside the salt mass (e.g., from an overlying aquifer) (Roedder, 1984a).

Some salt inclusions trap gases. The compositions and pressures of these gases can provide information on the formation of the salt, later changes to the salt, and the potential for gas movement through the salt (Siemann and Ellendorff, 2001). Gas inclusions in salt contain varying ratios of methane, carbon dioxide, and nitrogen and can be under pressures up to several hundred atmospheres (Roedder, 1984a). The origins of these gases remain a subject of debate and likely vary from site to site depending on the local occurrence of associated hydrocarbons, magmatic processes, or other sources. A large number of gas inclusions distributed throughout a volume of salt under sufficiently high pressure produce considerable stress and can result in mechanical failure of the salt mass when confining pressures are removed by mining. Large masses of salt have been displaced by such events, in some cases killing miners nearby (Hoy, et al., 1962).

Heat generated by waste emplaced in the salt will produce a thermal gradient that will vary in space and time as a complex function of numerous variables. Fluid inclusions in the salt will be affected by such a thermal gradient because salt solubility is a function of temperature. Generally, there will be a tendency for salt dissolution on the warmer side of the inclusion and precipitation on the cooler side. The result is that the fluid inclusions will tend to migrate up the thermal gradient toward the waste (Roedder, 1984b). Experiments by the U.S. Geological Survey using salt crystals from the WIPP site showed that in a thermal gradient of 1.5 °C/cm [6.9 °F/in] maintained for up to 10 days, rates of movement of fluid inclusions varied from 1.2 to 5.4 cm/yr [0.47 to 2.1 in/yr] (Roedder and Belkin, 1980). This effective migration of brine and its potential subsequent accumulation at the hot container surface could be a detrimental factor for container integrity because of the corrosive nature of the brine. The brine chemical composition and volume as well the container material would determine the type and extent of corrosion that could occur. Because salt formations have physical and chemical properties very different from those of other rock types, such as granite and argillite, other processes may occur that could affect the susceptibility of the container material to corrosion and will need to be considered.

## 4.2 Range of Brine Compositions

Published literature provides brine composition data. For example, Molecke (1983) listed the ionic compositions of 18 brines used by different investigators. Molecke (1983) divided these brines into two groups, "A-like" and "B-like" brines, based primarily on high NaCl or high Na-Mg-K-Cl content. The A-like and B-like brines have compositions similar to WIPP Brine A and Brine B, respectively, which Molecke (1976) originally classified to standardize brine compositions for use in waste form leaching and corrosion testing. Brine A is a high Na-Mg-K chloride brine representative of brines that have interacted with potassium and magnesium minerals and potentially could intrude into a waste facility in bedded salt. Brine A is similar in composition to brines found in small fluid inclusions in rock salt. Brine B is a near-saturated, predominantly NaCl brine representative of brines potentially intruding into either a domed salt repository or into a relatively pure bedded halite (Molecke, 1983). Although Brines A and B originally were defined as WIPP site-specific brines, their applicability has been broadened for use as generic test brines. Compositions of A-like and B-like brines Molecke (1983) compiled are listed in Tables 4-1 and 4-2, respectively.

The distinction between A-like and B-like brines was considered important because some experimental data indicated A-like brines cause a higher uniform corrosion rate of various alloys compared to B-like brines or seawater (Molecke, 1983). The higher corrosivity of A-like brines has been ascribed to their much higher magnesium concentration compared to B-like brines and the resulting lower solution pH when magnesium oxysulfates are precipitated or when  $\text{MgCl}_2$  in brine reacts with  $\text{SiO}_2$  (e.g., from the waste form or backfill). For example, the reaction according to Molecke (1983) is



However, there is no detailed study that shows  $\text{Mg}^{2+}$  concentration is the only major factor determining brine corrosivity.

Jain (1986) stated that because a brine is expected to be in equilibrium with the surrounding rock salt, its composition can be expected to have as wide a range as the salt mineralogy itself and it would not be possible to identify a unique brine composition that could represent all salt repositories. Consequently, some investigators have suggested the use of limiting compositions of brines corresponding to two scenarios of brine migration to the waste package (Jain, 1986). In the first scenario, external water enters and floods the repository to form dissolution brines expected to have a B-like brine composition. In the second scenario, existing brine inclusions migrate to the waste package due to a thermal gradient. Molecke (1983) suggested that inclusion brines will have an A-like brine composition. Although inclusion brines typically have a higher  $\text{Mg}^{2+}$  concentration than dissolution brines, exceptions to this observation are possible if the salt contains higher than normal amounts of magnesium compounds. Because salt compositions may vary between different sites, brine compositions need to be determined on a site-specific basis.

In addition to brine composition, the redox (reduction–oxidation) potential of the near-field environment also can be an important determinant of waste container corrosion, waste form degradation, and radionuclide solubility and transport. Generally, multivalent radioelements (e.g., uranium, plutonium, and neptunium) have lower solubilities and higher sorption on geologic media when in their lower oxidation states. However, salt formation by itself generally does not provide reducing environments and a HLW salt repository would be expected to have

**Table 4-1. Ionic Composition (mg/L) of “A-Like” Brines\***

“A-Like” Brines							Other Brines†		
Ion	(WIPP‡/ Generic Brine A (±3%)	WIPP‡ Inclusion No. 1 (Preliminary)	WIPP‡ Inclusion No. 2 (Preliminary)	MCC Brine	Quinare Brine Q {±10%; 55 °C [131 °F]}	USGS§ NBT-6a (±10%)	Saturated NaCl {20 °C [68 °F]}	Saturated NaCl {100 °C [212 °F]}	Sea Water
Na <sup>+</sup>	42,000	63,000 ± 5,000	32,000 ± 1,100	35,4000	6,500	27,000	142,000	154,000	10,651
K <sup>+</sup>	30,000	8,700 ± 500	6,800 ± 200	25,300	29,000	35,000	—	—	380
Mg <sup>++</sup>	35,000	23,000 ± 2,000	40,000 ± 1,400	29,600	85,000	33,000	—	—	1,272
Ca <sup>++</sup>	600	210 ± 20	150 ± 10	—	—	47,000	—	—	400
Sr <sup>++</sup>	5	—	—	—	—	—	—	—	13
Zn <sup>++</sup>	—	—	—	—	—	—	—	—	—
Li <sup>+</sup>	20	—	—	—	—	—	—	—	—
Rb <sup>+</sup>	20	—	—	—	—	—	—	—	—
Cs <sup>+</sup>	1	—	—	—	—	—	—	—	—
Fe <sup>+++</sup>	2	—	—	—	—	—	—	—	—
Cl <sup>-</sup>	190,000	160,000 ± 9,000	160,000 ± 5,000	164,000	270,000	250,000	218,000	237,000	18,980
SO <sub>4</sub> <sup>2-</sup>	3,500	13,200 ± 2,600	13,200 ± 2,600	—	13,000	—	—	—	884
B (as BO <sub>3</sub> <sup>3-</sup> )	1,200	—	—	—	—	—	—	—	—
HCO <sub>3</sub> <sup>-</sup>	700	—	—	—	—	—	—	—	146
Br <sup>-</sup>	400	—	—	—	—	—	—	—	65
I <sup>-</sup>	10	—	—	—	—	—	—	—	0.05
F <sup>-</sup>	—	—	—	—	—	—	—	—	—
pH	6.5	—	—	6.5	—	—	—	—	8.1

Blank cells indicate no data measurement reported.  
 \*Molecke, M.A. “A Comparison of Brines Relevant to Nuclear-Waste Experimentation.” SAND83-0516. Albuquerque, New Mexico: Sandia National Laboratories. 1983.  
 †The composition of other brines is also shown for comparison.  
 ‡ Waste Isolation Pilot Plant  
 §U.S. Geological Survey

4-4

**Table 4-2. Ionic Composition (mg/L) of “B-Like” Brines\***

Ion	(WIPP†/ Generic) Brine B (± 3%)	ONWI Composite Permian P	Equilibrated Permian P No. 2	Pretest PNNL‡- SNL§	Brine- Backfill PNNL‡- SNL§	Posttest PNNL‡- SNL§	Flow WIPP†-12	Downhole WIPP†-12	Flow ERDA-6	Downhole ERDA-6
Na <sup>+</sup>	115,000	123,460	123,000	159,000	155,000	119,000	114,000	140,000	112,000	140,000
K <sup>+</sup>	15	39	39	2,550	2,370	5,000	3,100	3,200	3,800	4,800
Mg <sup>++</sup>	10	134	122	409	463	158	1,700	1,400	450	270
Ca <sup>++</sup>	900	1,560	1,100	370	695	267	410	380	490	360
Sr <sup>++</sup>	15	35	35	18	43	34	15	—	18	—
Zn <sup>++</sup>	—	7.8	8	<2.5	—	199	0.5	—	0.6	—
Li <sup>+</sup>	20	—	—	—	—	—	220	210	240	205
Rb <sup>+</sup>	20	—	—	—	—	—	—	—	—	—
Cs <sup>+</sup>	1	—	—	—	—	64	—	—	—	—
Fe <sup>+++</sup>	2	—	—	<5	—	55	3.6	6.3	3.6	5.7
Cl <sup>-</sup>	175,000	191,380	191,000	190,000	231,000	197,000	160,000	180,000	170,000	180,000
SO <sub>4</sub> <sup>2-</sup>	3,500	3,197	1,910	2,086	3,237	16,300	17,000	18,000	16,000	14,000
B(as BO <sub>3</sub> <sup>3-</sup> )	1,200	—	—	16	11	1,280	1,200*	960*	680*	740*
HCO <sub>3</sub> <sup>-</sup>	700	30	23	7.8	15	0	2,600	2,400	2,600	1,800
Br <sup>-</sup>	400	32	24	—	—	—	430	460	880	720
I <sup>-</sup>	10	—	—	—	—	—	—	—	—	—
F <sup>-</sup>	—	1.1	1.0	37	37	0	4.3	—	1.7	—
pH	6.5	7.055	—	6.9	6.8	3.8	7.17	7.76	6.42	7.02
Field Eh (mV)	—	—	—	—	—	—	-211	—	-152	—

Values = reported as B, but assumed to actually be BO<sub>3</sub><sup>3-</sup>. — = below measureable detection limits, blank cells indicate no data measurement reported.

\*Molecke, M.A. “A Comparison of Brines Relevant to Nuclear-Waste Experimentation.” SAND83-0516. Albuquerque, New Mexico: Sandia National Laboratories. 1983.

† Waste Isolation Pilot Plant

‡ Pacific Northwest National Laboratory

§ Sandia National Laboratories

reducing near-field conditions only if reducing materials, such as steel, are sufficiently reacted with brine (Hansen and Leigh, 2011).

### 4.3 Temperature Effect on Brine Composition

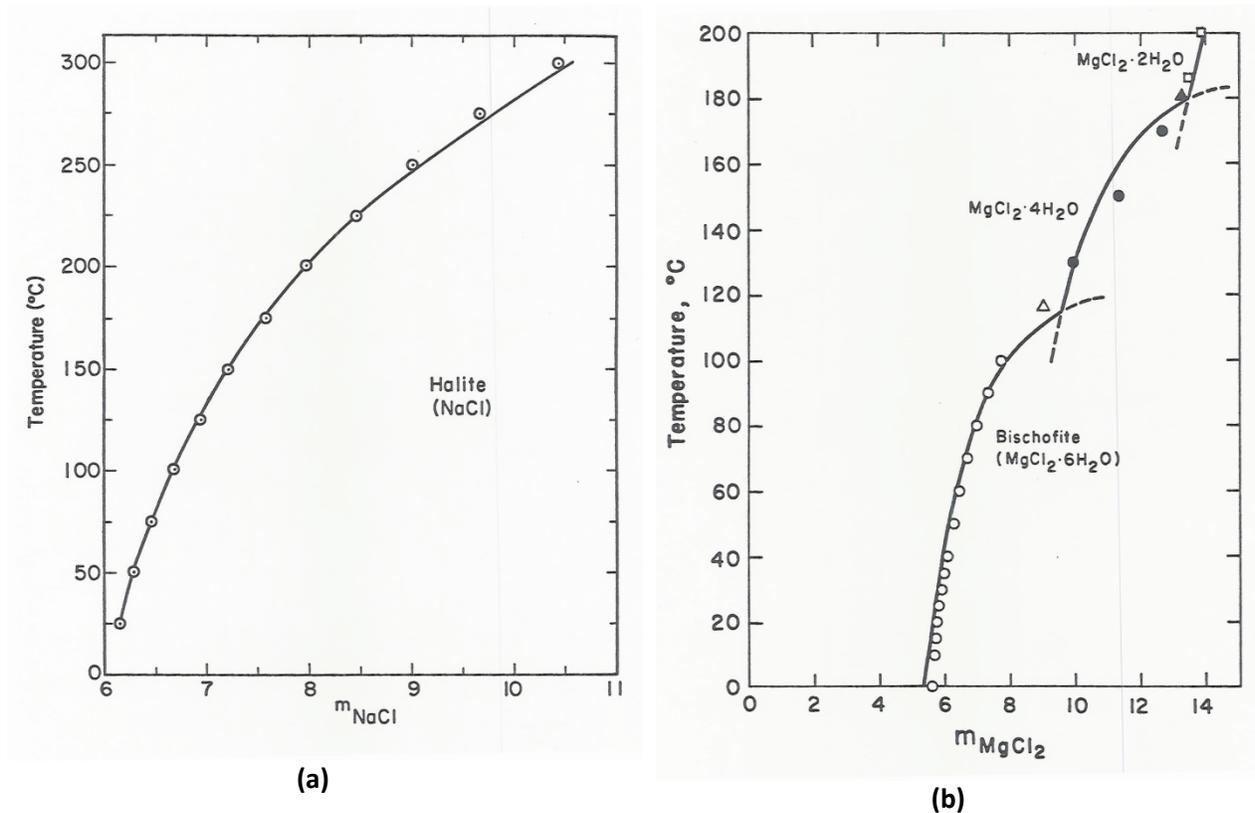
The brine compositions listed in Tables 4-1 and 4-2 are ionic concentrations measured at room temperature. However, the waste package temperatures will be elevated during the first several hundred years after repository closure. Salt solubility mostly increases with increasing temperature. Thus, brines in contact with hot waste packages will have higher ionic concentrations and likely would be more corrosive than brines at ambient conditions. On the other hand, the concentration of oxygen, which is an important species in corrosion reactions, decreases with increasing ionic concentration. If oxygen concentration is decreased significantly due to the higher ionic concentration, the corrosion rate may not be higher than at ambient temperatures.

Examples of increasing solubility with temperature are shown in Figure 4-1 for NaCl and MgCl<sub>2</sub> single salts. Note that salt solubility is more complex in systems with two or more salts present than in single salt systems due to salting out effects. However, thermodynamic models are available that can be used to calculate solubilities in multicomponent salt systems at various temperatures (e.g., Pabalan and Pitzer, 1991; Moller, 1988; Gruskiewicz, et al., 2007). Examples of calculated salt solubility as a function of temperature in two- and three-salt systems are shown in Figures 4-2 and 4-3. Thermodynamic calculations could aid in developing brine recipes for use in elevated temperature testing of waste form and waste container performance.

Temperature can affect brine chemistry in another manner. Uerpmann and Jockwer (1982) and Jockwer (1984) showed that the gases H<sub>2</sub>S, HCl, CO<sub>2</sub>, and SO<sub>2</sub> were released from natural salt from the Asse mine in Germany upon heating to 350 °C [662 °F]. Pederson (1984) reported the evolution of HCl, SO<sub>2</sub>, CO<sub>2</sub>, and H<sub>2</sub>S upon heating of Palo Duro and Paradox Basin rock salt to 250 °C [482 °F]. Impurities within the salt apparently contain one or more thermally unstable, acidic components. These components can volatilize during heating and increase the alkalinity of residual brines. For example, pH of brines prepared by dissolving Permian Basin salt samples that were annealed to progressively higher temperature {up to 167 °C [333 °F]} increased from near neutral to approximately 10 (Panno and Soo, 1984). In an open repository system, the acidic gases would escape and leave potentially alkaline-producing salt. However, in a sealed system, the gases may accumulate near the waste containers and eventually form acidic brines.

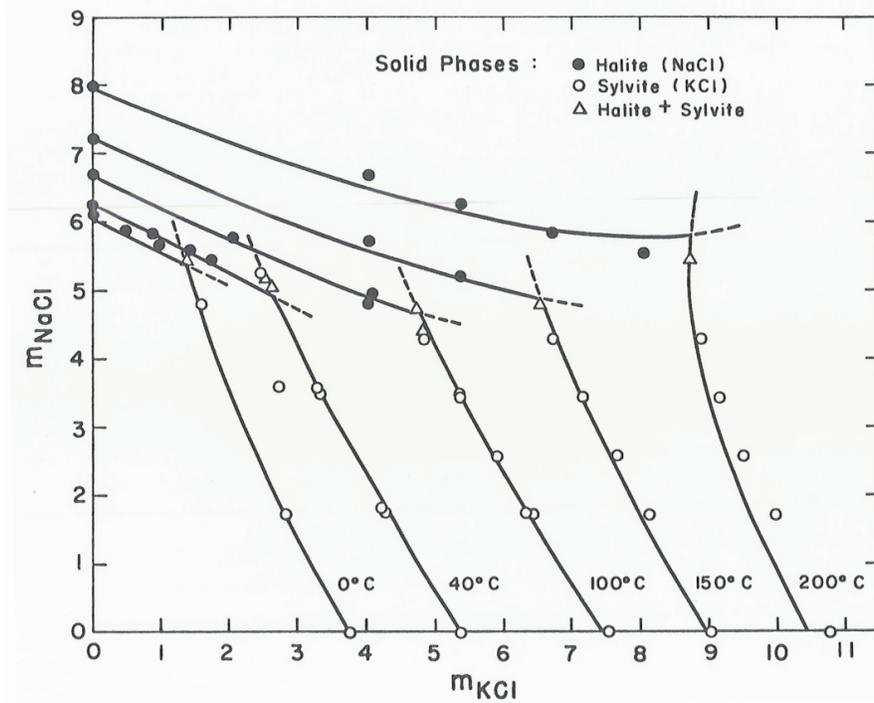
### 4.4 Radiation Effect on Brine Chemistry

Radiation can interact with the brine, change the brine chemistry, and affect corrosion processes. Jenks (1972) and Jenks and Walton (1981) reviewed published studies on the effect of radiation on simple salt solutions. Gamma radiation is likely to have a dominant effect on near-field conditions compared to particle radiation ( $\alpha$ ,  $\beta$ , and neutron) because the former is more penetrating than the latter. Although gamma radiation reaction with brine is complex, the major reaction is the decomposition of water into hydrogen and oxygen, accompanied by the production of the radicals e<sup>-</sup>(aq), H, OH, and HO<sub>2</sub> (or an ionic form of the radicals), molecular hydrogen (H<sub>2</sub>), and molecular hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>). The radicals e<sup>-</sup>(aq) and H are reducing radicals because they bring about reduction in inorganic solutes. Conversely, OH, HO<sub>2</sub>, and H<sub>2</sub>O<sub>2</sub> are oxidizing products because of their ability to oxidize inorganic solutes. The generation of several oxidizing and reducing ions and radicals can change the corrosion rates or

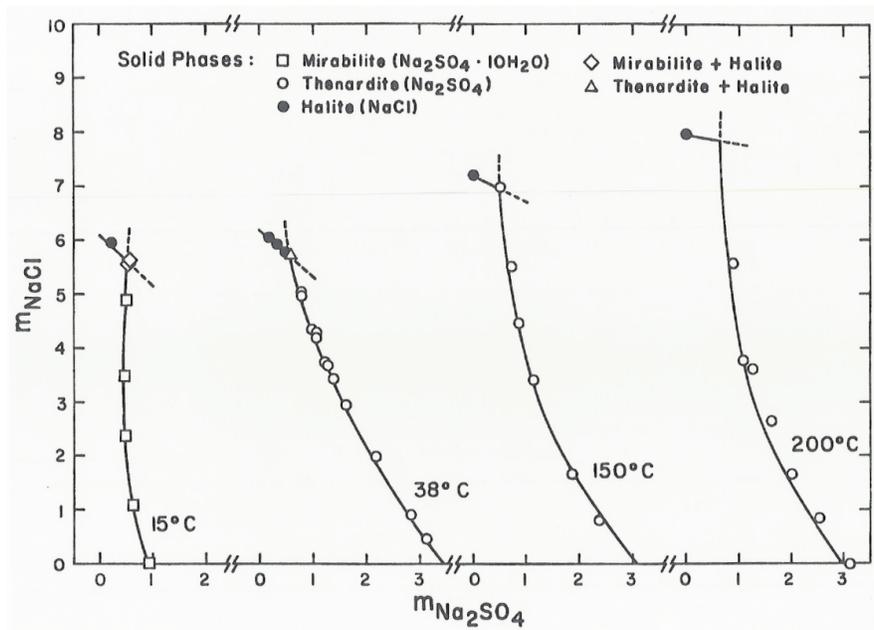


**Figure 4-1. Calculated Solubility of (a) NaCl and (b) MgCl<sub>2</sub> Salts in Water (Solid Line) Compared to Experimental Data (Symbols). Reprinted From Pabalan and Pitzer (1987) With Permission From Elsevier.**

mechanism occurring at the brine–waste package interface. Literature data reviewed by Jenks (1972) and Jenks and Walton (1981) indicate that brine radiolysis can change the pH and Eh of the liquid phase and generate gases (primarily hydrogen). Irradiation of brine solutions produces large amounts of gas together with other stable species. The principal reaction that produces gases during radiolysis is the decomposition of the water molecule into hydrogen and oxygen. The presence of cations and anions in brine may affect the final yield of gases by scavenging the intermediate species. In general, gamma radiation appears to drive brines toward neutral pH. However, the extent of pH change may depend on several parameters, such as composition, temperature, dose, type of radiation, and salt mineralogy. The degree to which these parameters change pH needs further study. In addition, little information is available on the effect of radiation on brine Eh as well as on the effect on corrosion of the highly reactive transient species produced by irradiation.

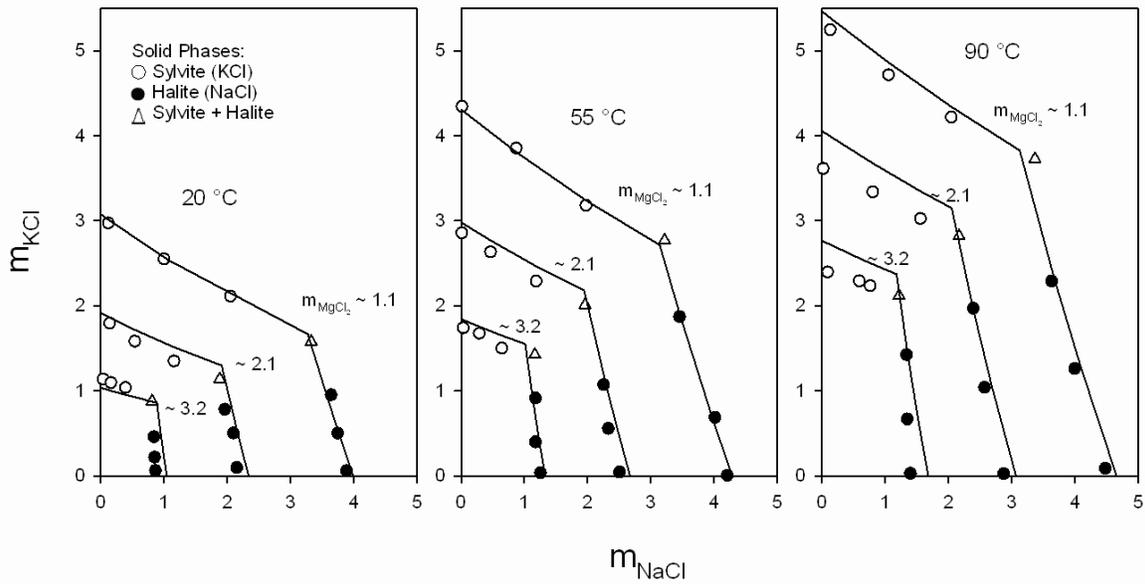


(a)



(b)

**Figure 4-2. Calculated Salt Solubility (Solid Lines) As a Function of Temperature in Two-Salt Mixtures of (a) NaCl+KCl and (b) NaCl+Na<sub>2</sub>SO<sub>4</sub> Compared to Experimental Data (Symbols). Reprinted From Pabalan and Pitzer (1987) With Permission From Elsevier.**



**Figure 4-3. Calculated Salt Solubility (Solid Lines) As a Function of Temperature in Three-Salt Mixtures of NaCl+KCl+MgCl<sub>2</sub> Compared to Experimental Data (Symbols). Reprinted From Pabalan and Pitzer (1987) With Permission From Elsevier.**

## 5 WASTE PACKAGE CORROSION

One of the main threats to the integrity of the waste canister is the aqueous corrosion of the outer canister material. Rates of corrosion depend on the geochemical environment and the type of material used in the waste package. Presently, none of the existing programs for evaluating salt formations for geologic disposal of heat-generating waste has formally specified a waste package design or canister material. Additionally, there is no clear consensus in the literature as to what canister materials would be preferable for a salt repository application. Generally, two basic types of waste package applications have been considered and are discussed in this chapter. The first is a corrosion-resistant material application, in which an outer corrosion-resistant overpack is included in the design to reduce corrosion rates as much as possible to limit releases until either radioactive decay has significantly diminished the source or beyond the period of geologic stability. The second is a corrosion-allowance material application, in which corrosion is allowed to occur, and the primary function of the canister is to provide sufficient shielding and waste isolation to contain the fuel through emplacement operations and through the postclosure thermal period until the formation can be relied on to provide effective waste isolation. Potential container materials and applicable corrosion processes for the two waste package applications are discussed in the following two sections.

### 5.1 Corrosion-Resistant Material Application

Various materials have been evaluated for use as a thin, corrosion-resistant overpack in a salt repository. Testing was conducted at WIPP to evaluate the corrosivity of defense HLW packages in salt conditions (Molecke, 1984). In this study by Molecke (1984), canisters were fabricated from Class 12 Titanium material. In addition, canisters fabricated of mild steel had an overpack of Class 12 Titanium. Helie and Plante (1985) evaluated the corrosivity of not only the titanium alloy with 0.2 percent palladium but also nickel-chromium alloys including Alloy 625, C276, and C4 in a salt environment. The corrosion resistance of the canister will depend upon the environment and alloy composition, but the main mechanisms of degradation include general corrosion, localized corrosion, hydrogen embrittlement, and radiation effects.

General corrosion occurs when the overall surface of a material corrodes at a uniform rate. General corrosion is not typically considered a degradation mechanism of concern for this application, because many of the corrosion-resistant materials form a stable and protective film. For example, titanium has been shown to have low corrosion rates in oxygen containing saltwater with a pH of roughly 4.5 and temperature of 100 and 130 °C [212 and 266 °F] (Magnani, 1983). However, the uniform corrosion of titanium could be an issue depending upon the chemical composition or crystallographic microstructure of the alloy. Ruppen, et al. (1983) showed that Class 12 Titanium could have a 100 percent increase in the corrosion rate by sensitizing the material in a particular temperature range. Similarly, Ruppen, et al. (1983) showed that by increasing the iron content from 0.05 percent to 0.37 percent, the corrosion rate increased roughly 50 to 100 percent. Therefore, examination of the microstructure that can arise from welding processes and also the composition ranges of alloys need to be understood for application in a salt repository. However, general corrosion is not typically an aging mechanism of concern for corrosion-resistant materials.

The main issue with corrosion-resistant materials is localized corrosion, such as crevice and pitting corrosion. Localized corrosion generally refers to a process where corrosion occurs at discrete sites, in contrast to general corrosion, which uniformly thins the entire surface of a material. Materials with a protective passive film are more likely to be susceptible to localized

corrosion rather than general corrosion. Both titanium and nickel-chromium alloys are known to undergo crevice corrosion at higher temperatures (Magnani, 1983). Ahn, et al. (1983) explain that crevice-type environments are expected to form in the repository between the container and the backfill material or metallic emplacement sleeves. The study by Schwarzkopf, et al. (1992) examined both Titanium 99.8-Pd and Hastelloy C4 in either a sodium chloride brine or magnesium chloride brine. Localized corrosion was not observed on the titanium alloy canisters after an 18-month exposure time. However, the Hastelloy C4, while resistant in sodium chloride solution, had a large number of localized ruptures in magnesium chloride brine when exposed to temperatures between 150 and 200 °C [302 and 392 °F]. Ahn, et al. (1983) evaluated Class 12 Titanium in a simulated brine and observed crevice corrosion at 150 °C [302 °F].

Another form of localized corrosion is known as stress corrosion cracking. Stress corrosion cracking is the cracking of a material when three conditions are met: a susceptible material, stressed state, and a susceptible environment (Toshima and Ikeno, 2000; Huizinga, et al., 2005; National Aeronautics and Space Administration, 1971). Of the possible conditions in a salt repository, stress corrosion cracking may occur depending upon the exact material and environmental conditions. Slow strain rate tests conducted on Class 12 Titanium showed that this material was not susceptible to stress corrosion cracking up to a temperature of 250 °C [482 °F]. However, other types of titanium alloys have been shown to be susceptible to stress corrosion cracking at temperatures as low as room temperature (Schutz, 2005). Therefore, it is important to evaluate the potential canister material in the salt environment to ensure cracking will not occur.

In conjunction with stress corrosion cracking, hydrogen embrittlement is also an environmental degradation. Hydrogen embrittlement is a degradation mechanism that can lead to higher susceptibility in cracking, including stress corrosion cracking. Hydrogen cracking is especially a concern for titanium alloys because the hydrogen can precipitate as titanium hydrides, which embrittles the material. Hydrogen can be formed by the corrosion of the metallic materials or through radiolysis of the brine or groundwater. Ruppen, et al. (1983) examined hydrogen embrittlement of Class 12 Titanium and identified that this is a credible issue when the hydrogen concentrations reach 200–300 ppmw, but will depend upon the titanium alloy chemical composition. This is in agreement with the results Moody and Robinson (1983) observed. Hydrogen embrittlement in titanium is an even greater concern in crevice regions because of the lower pH and higher corrosion rates, which could lead to higher hydrogen production (Ahn, et al., 1983).

The final degradation issue is radiation effects. While radiation is not expected to modify the metal canister material physically, it is expected to modify the surrounding geochemical environment, which could lead to higher rates of degradation. Magnani (1983) stated that Class 12 Titanium showed a twofold increase in the corrosion rate when exposed to irradiated brine and seawater. Shoemith, et al. (1992) evaluated the stress corrosion cracking that may occur depending upon the exact material and environmental condition effects of radiation on waste containers for different materials. For nickel-based alloys, they indicated an increase in localized corrosion leading to a weight loss of greater than 100 times for Hastelloy C4 in highly saline brines. The increase in corrosion rate is likely due to the formation of radiolytic oxidants.

## **5.2 Corrosion-Allowance Material Application**

The typical materials examined for use in a corrosion allowance application are various ferrous alloys, such as cast iron, cast steel, or wrought mild steel. During the 1980s, a variety

of studies examined the corrosion properties of ferrous alloys for use as a waste package material (Haberman and Frydrych, 1988; Hauser and Köster, 1985; Magnani, 1983). These studies examined general corrosion, localized corrosion, stress corrosion cracking, and radiation effects.

The main corrosion mechanism for ferrous alloys is general corrosion, which starts at a high rate and slowly decreases with time for most alloys. Haberman and Frydrych (1988) examined ASTM A216 steel exposed to brine solutions simulating salt repository conditions. In the case of brine solution with low magnesium at 150 °C [300 °F], the initial corrosion rate was roughly 0.05 mm/yr [0.002 in/yr], which decreased to 0.02 mm/yr [0.001 in/yr] after 3 months and then remained constant. In the high magnesium case at the same temperature, the corrosion rate increased to as high as 1.5 mm/yr [0.06 in/yr] and then slowly decreased with time. Temperature, in addition to time and chemical environment, also affects corrosion rate. Haberman and Frydrych (1988) showed that the corrosion rate of ASTM A216 steel increased exponentially between the temperatures of 100 and 200 °C [212 and 392 °F]. In addition to the environmental effects, the specific alloy chosen will also affect the general corrosion rate. Westerman and Pitman (1985) showed a range of corrosion rates for ASTM A216 cast steel, ductile cast iron, and wrought 1025 steel for brine solutions simulating conditions in the Permian basin.

Localized corrosion in the form of pitting or crevice corrosion occurs for ferrous alloys in different environments (McNeil, 1987). However, pitting corrosion may not occur in the repository environment, because the general corrosion rate is so high. Studies by Smailos, et al. (1992) and Schwarzkopf, et al. (1992) on the corrosion rate of ferrous-based alloys in salt repository conditions only showed general corrosion occurring on the surface of the materials.

Localized corrosion in the form of stress corrosion cracking is not expected to occur for these ferrous alloys in salt repository conditions. A study by Pitman (1987) examined ASTM A216 cast steel in a 3-month salt exposure test using stress corrosion cracking U-bend specimens. The samples were held at either 90 or 150 °C [194 or 302 °F], and none of them were observed to have cracked. The results of this study suggested that ASTM A216 cast steel is not susceptible to stress corrosion cracking under the test conditions examined. Westerman, et al. (1984) conducted another study to evaluate the stress corrosion cracking susceptibility of ferrous-based alloys in brines representing rock salt solutions. Similar to the Pitman study, none of the specimens examined showed any evidence of either transgranular or intergranular fracturing.

The effect of radiation is the same as for the corrosion-resistant application, in that radiation's main effect is to increase the corrosivity. Westerman, et al. (1984) examined the irradiation effect on ferrous alloys by conducting corrosion studies in a Co-60 irradiation facility. The results of this study indicated that the corrosion rate in the irradiated tests was much higher than in the un-irradiated tests. Similarly, Shoosmith, et al. (1992) indicated that above certain radiation levels, the corrosion of ferrous alloys can increase as much as 15 times more than in the un-irradiated condition.

## 6 SUMMARY AND CONCLUSIONS

Deep burial of HLW in salt formations is currently being considered as a potential solution for disposal of the nation's inventory of spent nuclear fuel and other heat-generating radioactive waste. The most often cited favorable attributes of salt disposal are the low permeability of undisturbed salt and the potential for fractures and openings in salt to self-seal. Experience and knowledge gained from the WIPP site and from petroleum storage in salt domes provide evidence that salt formations are capable of isolating large volumes of environmentally hazardous substances under isothermal conditions. In the United States, there are several locations where thick, laterally extensive salt formations are accessible. As discussed in the preceding chapters, however, there is great variability in the physical and mechanical properties of salt formations and not all salt formations may be suitable for permanent HLW disposal. In the introduction, four key site-specific processes and characteristics were identified as the main components that facilitate waste isolation in a salt repository system. These four components are revisited next with summaries of insights obtained from the foregoing discipline-based discussions in Chapters 2–5.

1. Permeability Barrier Effectiveness. Available information and observations indicate that stress changes due to excavated openings and the introduction of a significant heat source will result in a deformation damage zone around the repository. This deformation zone would enhance permeability in the near field surrounding the emplacement drifts. Due to creep processes, the damaged zone will tend to gradually heal over time, but permeability may remain somewhat higher than the undisturbed salt formation because of the introduction of additional porosity to the system. The volume of the formation that would be affected by enhanced permeability cannot be estimated without knowledge of site-specific rock properties and design information. Therefore, candidate sites will need to be evaluated for a specific repository design and heat load to ensure the formation is thick enough to contain the damage zone with sufficient undamaged formation to maintain waste isolation. Candidate sites also need to be evaluated for hydrologic characteristics of the far field, including the potential for erosion-, uplift-, or climate-induced changes in groundwater circulation patterns that could lead to shallow, lateral, or deep dissolution of the salt formation.
2. Brine/Water Mobility in the Repository Near Field. As discussed previously, deformation due to excavation and heat will likely produce a zone of enhanced permeability surrounding the near field. This zone could facilitate movement or circulation of fluids and vapor around the waste canisters. Salt formations suitable for geologic disposal generally will have low water content, mainly in the form of fluid inclusions entrapped in the salt at the time of formation. Near-field thermal and hydrological processes can drive movement of fluid inclusions toward the heat source until they reach some type of fracture or open space where they could become more mobile. During the period of high thermal load, fluids entering fractures or open space near waste packages would likely be driven away in vapor form, potentially condensing in cooler areas farther from the heat source. As the waste cools and the deformation zone begins to heal over time, the long-term migration of fluid inclusions into the damage zone may result in a zone of relatively higher—but still fairly low—fluid content surrounding the waste packages. Barring intrusion or external disturbance, there is little threat of this fluid migrating out of the near field, so long as the formation is thick enough to contain the damage zone with a sufficient undamaged formation to maintain waste isolation.

3. Waste Package Integrity. Corrosion processes are the primary threat to waste package integrity in a salt repository system. Titanium or nickel-chromium alloys are potential materials that would be used for canister overpacks in a salt repository. Both of these materials are known to undergo crevice corrosion at high temperatures. Stress corrosion cracking may also occur, depending upon the exact material and environmental conditions. Corrosion rates for both titanium and nickel-based alloys have also been shown to increase in the presence of concentrated brine. Uniform corrosion of titanium could be an issue, depending on the chemical composition or crystallographic microstructure of the alloy. Expected lifetimes of waste packages in a salt repository will depend on the particular alloy chosen and the site-specific chemical environment. Waste package integrity may also be affected by external mechanical forces. Generally, waste packages can be designed to withstand anticipated stresses, but when combined with weakening due to corrosion, mechanical stress may accelerate waste package degradation and crack propagation.
  
4. Radionuclide Mobility. In the event of a waste package breach, radionuclide mobility would require the presence of sufficient liquid to facilitate dissolution of waste and transport of the waste outside of the package. Therefore, radionuclide dissolution and transport outside of the waste package may be greatly impeded by the limited water content of salt and, if localized corrosion or stress corrosion cracking are the failure modes, by limited breach area through which solutes could exit the waste package. Radionuclide solubility will be lower for concentrated brine compared to less saline groundwater. Chemically reducing environments are also known to greatly reduce solubilities of many radionuclides; however, a salt formation by itself generally does not provide reducing environments. A salt repository would be expected to have reducing conditions only if reducing materials, such as steel, are sufficiently reacted with brine to consume available oxygen. If radionuclides are able to exit the waste package, they may be relatively mobile within the near field due to damage-enhanced permeability and the potential heat-driven increase in fluid content. The sorption potential of salt is very low. However, at the very low permeabilities associated with undisturbed salt formations outside the near field, there is essentially no advective movement of water, so radionuclide transport away from the repository is limited to diffusion in whatever brine may be present in interconnected pathways (e.g., along grain boundaries) in the salt. In the absence of an intrusion, external disturbance, or undetected permeable feature, the limited mobility in salt could effectively halt radionuclide transport. In the event that radionuclides could exit the salt formation, site-specific transport properties of the far field would determine radionuclide mobility.

A recurring theme in these insights is that the salt formation itself appears to be the principal barrier that facilitates waste isolation. During the process of site selection, it will be important to evaluate whether the formation is of suitable integrity and thickness to ensure that the formation of a dilation damage zone around the waste emplacement will not propagate through the formation. Reliable site-specific characterization data and design information are needed to conduct such an analysis. Additionally, models capable of considering the complex deformation processes that affect the extent of the dilation damage zone will need to be identified or developed and validated. Recommended future work also includes (i) understanding impacts of potential disturbances (e.g., seismic activity); (ii) evaluating stylized scenarios (e.g., human intrusion) affecting the barrier capability of the salt formation; and (iii) a detailed evaluation of all available FEPs for radioactive waste disposal in salt formations.

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