

BRINE MIGRATION IN SALT

TOPICAL REPORT

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by

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## I. INTRODUCTION

In order to dispose of high-level radioactive waste (HLW), the Department of Energy (DOE) is mandated to build a deep geologic repository. One of the proposed rock types in which this repository may be built is salt (bedded or domal). A favorable aspect of salt is that it has low permeability, thus inhibiting the flow of groundwater. Groundwater is the most important agent for corrosion of the waste package, solubilization of the waste form, and transport of the waste to the accessible environment. Salt has been proposed as an ideal host rock because it is generally considered an anhydrous medium based on the general formula NaCl. However, salt deposits do contain water that was either trapped during deposition of the salt or emplaced into the salt during some secondary process. This water may be present in intercrystalline inclusions (e.g., brine pockets), intracrystalline inclusions, and hydrated or hydrous minerals. The concern with brine inclusions in salt is that, under a temperature and/or pressure gradient, they have been observed to migrate. The water can move either up or down a gradient, depending upon the nature of the inclusion. Therefore, despite the basic impermeability of salt, water may still reach the waste package and affect its performance under repository-induced conditions. Information obtained by study of the composition of inclusions in the salt can aid in the understanding of the depositional and diagenetic history of the salt. It is therefore important to understand the nature of these inclusions and their potential impact on the performance of a HLW repository. This report summarizes the work that has been completed to date on inclusions in salt and on brine migration and discusses uncertainties in predicting the effects of brine migration on repository performance.

## II. FLUIDS IN SALT

### Types of Fluids

As stated above, fluids in salt deposits are found in three settings: (1) intercrystalline inclusions; (2) intracrystalline inclusions; and (3) hydrated or hydrous minerals. Intercrystalline inclusions contain water found between the mineral crystals along grain boundaries and in fractures. These inclusions can contain large amounts of brine (i.e., brine pockets) and can be under high pressure. A brine pocket containing  $2.7 \times 10^6 \text{ m}^3$  of brine was encountered at the WIPP site (National Research Council, 1984). Most salt crystals contain some amounts of water within the crystal structure, i.e., intracrystalline inclusions. These inclusions are commonly between 2 and 250 microns in diameter, although inclusions of several millimeters in diameter have been found (Isherwood, 1979). Inclusion sizes and shapes can vary greatly within a single crystal, although the smaller the inclusion, the more cubic its shape tends to be. The amount of fluids present in salt depends on the crystallization history of the salt. The higher the growth rate of salt crystals precipitating out of solution, the greater the amount of fluid trapped

in inclusions (see Roedder, 1984, for a more complete discussion). Most salt, however, undergoes recrystallization during its diagenetic history. This recrystallization process may be isochemical but also may involve the loss and/or gain of fluids (Roedder, 1984). An example of such an effect is the diapirism of salt beds (i.e., the formation of salt domes). Salt domes generally contain less included water than bedded salts, often more than an order of magnitude less. This is postulated to be a result of the squeezing out of fluids, apparently along grain boundaries, as the salt is forced toward the surface (Baes and other, 1983).

Certain minerals commonly associated with salt deposits can contain significant amounts of water. The common minerals that make up the vast majority of salt deposits, halite, anhydrite and sylvite, contain no water in their crystal structures. Minerals such as carnallite, kieserite, gypsum, and polyhalite, however, do contain bound water and are commonly found in salt deposits. In addition, sedimentary units such as shales which contain clay minerals are always associated to some extent with salt deposits. Under repository-induced temperatures, the hydrous and hydrated minerals may become unstable and undergo dehydration reactions. The more "pure" the salt is, the less likely it is to contain significant amounts of water in hydrous and hydrated minerals.

#### Composition of Included Water

The total water in salt deposits may range from approximately 30 ppm in some dome salts (Knauth and Kumar, 1981) to several weight percent in some bedded salts (Roedder and Belkin, 1979a). Because of the small size of inclusions found in most salt deposits, little quantitative work has been done studying the composition of fluid inclusions. Most of the work has been done on the rare, large inclusions (cm-sized); many studies of the smaller inclusions (mm-sized) have been performed in the USSR (see Petrichenko, 1973). The data suggest that the fluids are Ca-Mg brines with varying amounts of  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{=}$ , and other trace constituents (Isherwood, 1979). The total dissolved solids (TDS) values range from 32 to 36 weight percent, depending on the Ca:Mg ratios (Clynne and Potter, 1977). Little is known about the identity and concentration of the trace constituents. Solid phases are found in some inclusions, including halite, sylvite, anhydrite, carnallite, epsomite, bischofite, and possibly cryptomelane (Isherwood, 1979; Roedder, 1984). A number of gas phases in fluid inclusions have been identified, including  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{H}_2$ ,  $\text{N}_2$ ,  $\text{CH}_4$ ,  $\text{C}_2\text{H}_6$ ,  $\text{H}_2\text{S}$ ,  $\text{O}_2$ , Ar, Xe, Ne, and He (Roedder, 1972). Oil globules have also been observed (Roedder and Blekin, 1979b). Inclusions formed during different periods of crystal growth and because of post-depositional processes can have dissimilar chemical compositions; inclusions in the same deposit or even adjacent to one another can differ significantly.

Little information is available concerning the redox and pH conditions of the fluids in inclusions. Quantitative data are limited to a few Russian studies.

The pH is generally acidic (between 3 and 6.5) while redox conditions (Eh was measured) varies considerably (Isherwood, 1979). Petrichenko (1973) observed that fluids from inclusions in salt domes tended to be reducing (Eh = 10 to -210 mv) while those in bedded salt tended to be more oxidizing (Eh > 100 mv).

Information from hydrogen and oxygen isotope studies generally show that the fluids are mixtures of seawater and local meteoric waters from a variety of possible sources, rather than seawater residues only (Beeunas and Knauth, 1983; see Roedder, 1984). Studies by Knauth and others (1980) and Knauth and Kumar (1983) show that some water from brine leaking in Weeks Island, Jefferson Island, and Avery Island salt mines in Louisiana is meteoric in origin.

### III. THEORY OF BRINE MIGRATION

The migration behavior of intracrystalline inclusions under a temperature gradient has been extensively studied (e.g., Anthony and Cline, 1971; Cline and Anthony, 1971; Geguzin and others, 1975; Olander and Machiels, 1979; Olander, Machiels, and Yagnik, 1980; Jenks and Claiborne, 1981; Pigford, 1981; Biggers and Dayton, 1982; Yagnik, 1983; Olander, 1984). The nature of the movement is dependent upon whether the inclusion is all-liquid or contains a gas phase.

When a temperature gradient is applied to an all-liquid inclusion, migration is driven by solubility differences; i.e., since the solubility of salt increases with temperature, salt will dissolve at the interface closest to the heat source (the "hot" face), molecularly and thermally diffuse through the brine, and precipitate at the interface furthest from the heat source (the "cold" face). Thus, through the dissolution-precipitation process, the inclusions will migrate toward the heat source. The processes that control migration velocity are the diffusional transport within the inclusion and the kinetics of interfacial mass transfer (i.e., the dissolution and precipitation within the inclusion) (Yagnik, 1983).

The presence of a gas phase within the inclusion will profoundly affect brine migration. The increase in vapor pressure of water over brine with temperature controls inclusion behavior. Water will evaporate at the hot side of the gas bubble and be transported to the cold side, condense, and dissolve salt until saturation is reached (Anthony and Cline, 1972). The condensed water in turn backflows to the hot side, carrying dissolved salt from the cold face of the inclusion to the hot face. This backflow effect overwhelms any solubility-driven salt flux if the gas volume fraction is > 0.1. Thus, the gas-liquid inclusion will migrate away from the heat source.

The behavior of intercrystalline inclusions under a temperature gradient is unknown. It has been postulated, however, that their migration is driven by pressure gradients (Jenks and Claiborne, 1981, Rothfuchs et al., 1984); i.e., intercrystalline inclusions will migrate toward areas of low pressure. This

phenomenon is observed in salt mines where water in the salt under lithostatic pressure leaks into the mine, which is under atmospheric pressure. The behavior of water released from hydrous and hydrated minerals in a salt deposit is also unknown. Clay minerals are commonly found in stringers or interbeds. Water liberated from clay minerals due to heating may preferentially migrate within interbeds. This released water may undergo significant migration in clastic zones in a manner not predicted by brine migration theory. In addition, released water encountering salt minerals will dissolve the salt until solubility limits are reached in the resulting brine. Thus, some brine migration may occur in this manner.

#### IV. EXPERIMENTAL STUDIES

##### Laboratory Studies

As stated above, almost all of the experimental studies on brine migration to date have been concerned with intracrystalline migration. Some of the pioneering laboratory work was done by T.R. Anthony and H.E. Cline (1971, 1972, 1974; Cline and Anthony, 1971, 1972). They studied thermal migration of both liquid droplets and biphasic vapor-liquid droplets in solids and through grain boundaries. They used high-purity KCl crystals and produced their own brine inclusions by drilling holes into the crystals, filling them with deionized water, and sealing them with wax to prevent evaporation. A temperature gradient was then applied and inclusion behavior observed. Initially, the large inclusions tended to break up into many smaller inclusions under the temperature gradient. As the inclusions began to migrate under the temperature gradient, Anthony and Cline (1971) observed that they changed shape, becoming more disk-like. Larger inclusions traveled faster than smaller ones and were observed to collide and coalesce with smaller ones as they migrated. The investigators noted that migration was heterogeneous and should be treated as such. They suggested that interfacial kinetics control inclusion migration occurrence and velocity and that there will be no migration below a certain critical inclusion size (Anthony and Cline, 1971).

In other studies, Anthony and Cline observed migration behavior at grain boundaries (Cline and Anthony, 1971) and biphasic vapor-liquid inclusion migration (Anthony and Cline, 1972). The investigators observed that the surface tension of a grain boundary tended to pull droplets that had passed through a boundary back towards the boundary plane under a temperature gradient. This effect was negated if the temperature gradient was increased. Anthony and Cline found that a vapor phase in an inclusion would cause it to move down a temperature gradient (see Section III) and that the nature of the vapor phase affects the velocity. If the gas phase is only water vapor, viscous gas flow controls transport of water through the vapor phase. If a significant "foreign" gas is present, diffusion controls the transport of water vapor through the gas phase and thus the migration velocity is significantly less.

Salt from the Hutchinson mine in Kansas was used to determine the effect of temperature on salt (Bradshaw and McClain, 1971). The investigators found that salt fractured with considerable violence (i.e., decrepitated) when heated to around 280°C, independent of the rate of heating. This violent fracturing was interpreted to be caused by the pressure of the heated brine. The investigators identified a number of studies of salt from other mines in which fracturing of salt under high temperatures was observed. Bradshaw and McClain (1971), calculated that 2 to 10 liters of brine released by fracturing would migrate to a waste disposal hole in 20 to 30 years in the Hutchinson mine. These values were supported by laboratory studies.

There is a substantial body of work by Soviet investigators on brine migration (Geguzin and Dzyuba, 1973a, 1973b, 1974; Geguzin and others, 1975; Sedletskii and others, 1973). Isherwood (1979) presents an annotated bibliography on these works.

Thermal experiments were conducted at Sandia National Laboratories on large salt blocks as a prelude for subsequent in situ experiments at the Waste Isolation Pilot Plant (WIPP). In the Salt Block I tests, a cylinder of salt one meter in diameter and one meter high was heated and various tests were performed (Duffey, 1979). Some water loss data were collected and are reported but not interpreted. The Salt Block II tests were specifically set up to observe brine migration in a salt block (Hohlfelder, 1980). The salt block used contained total volatiles (mainly water) of between 0.1 and 0.5 weight percent. The salt itself was 88% halite, 8% polyhalite, and 4% sylvite (Lambert, 1979). A maximum temperature of 200°C and a temperature gradient of 12°C/cm were used. The experiment ran for approximately 140 days. Only 111.4 grams of water were released (compared to between 2.2 to 9.0 kg of water initially), and 43% of that was released during the three-week-long period after the heater was turned off. In general, the greatest water releases were observed following decreases in heater power. The investigators suggest that "the thermochemical response of the salt, including the stress state and time-dependent stress relaxation, is an essential ingredient in predicting the measured water release rates" (Hohlfelder, 1980). The investigators suggest that when the heater power was decreased, the salt failed in tension and thus increased in permeability, resulting in an increase in water release. Similar results were found in additional tests at Sandia (Hohlfelder and Hadley, 1979).

Additional results from the Salt Block tests show that encrustations on the interior of the axial heater hole consisting of sodium, potassium, and magnesium chloride minerals (e.g., carnallite, bischofite, recrystallized polyhalite) not usually found in the host salt rock were formed (Lambert, 1979). Investigators also observed that, in general, only the larger inclusions crossed grain boundaries during thermal migration. However, the larger inclusions were also observed to stop at least 7 cm from the heater hole; smaller inclusions were predominant closer in, with a vapor phase commonly present in the inclusion within 4 cm of the heater hole (Lambert, 1979). This may be due to a breakup of the larger inclusions and boiling of the fluids close to the heater.

Roedder and Belkin (1979a, 1980) also performed laboratory studies on WIPP salt. The investigators found that the fluids in many large inclusions ( $> 100 \mu\text{m}$ ) were released by decrepitation at  $250^\circ\text{C}$  in several days (Roedder and Belkin, 1979a). This decrepitation caused some fracturing and crumbling in the 100 gram pieces of core used. At grain boundaries, larger inclusions were found to cross the boundaries while smaller ones would not (if the boundary was tight). In a subsequent experiment, approximately one centimeter cubed blocks of salt cut from portions of single crystals were heated and intracrystalline inclusion behavior observed (Roedder and Belkin, 1980). Only relatively small inclusions ( $< 10^9 \mu\text{m}^3$ ) were present. The investigators observed that migration rates could differ by a factor of three for different samples from the same core or even from different parts of the same piece of core.

Investigators at the University of California-Berkeley have performed considerable experimental work on brine migration (e.g., Olander and others, 1980, 1982; Yagnik, 1983; Olander, 1984). The investigators used a microscope with a hot stage and observed all-liquid and gas-liquid inclusions in both NaCl and KCl (Olander and others, 1980). Two of their observations were that very large inclusions tended to break up into smaller ones under a temperature gradient and that migration velocities were affected by the number of dislocations present in the salt crystal (Olander and others, 1980; Yagnik, 1983). Additional studies showed that interfacial kinetics (i.e., the processes affecting movement at grain boundaries) were rate-controlling for inclusion migration velocity except for large inclusions. Large inclusions intersect enough dislocations so that the kinetic resistance to dissolution is negligible and diffusion is the rate-controlling process (Olander and others, 1982; Yagnik, 1983). The effect of stress was also observed. Migration velocities rose abruptly under an increased axial loading and increased with time. The velocities did not return to initial values after removal of the load, indicating an inelastic property. A change in brine composition (i.e.,  $\text{MgCl}_2$  and  $\text{SrCl}_2$  solutions) did not affect migration velocity (Yagnik, 1983).

Gas-liquid inclusions were observed to behave similarly to all-liquid inclusions except for the direction of migration. Investigators also observed inclusion behavior at grain boundaries. Inclusions migrated up or down the temperature gradient (depending upon the inclusion make-up), but in an indirect, two-dimensional manner (Yagnik, 1983). This was interpreted to be due to the distortion in the solid lattice at the grain boundary and the fact that a grain boundary is an area of minimum energy which would promote inclusion migration along the grain boundary.

Biggers and Dayton (1982) performed brine migration experiments on polycrystalline salt samples that were hot-pressed to control the microstructures. Salt samples were put in contact with a brine source and a temperature gradient was induced. Migration velocities were estimated by the timed appearance of brine at the hot face or by determining the penetration distance by migration artifacts in the microstructure. Visual observation was precluded due to the nature of the experimental apparatus. A significant result of the study is that migration velocities in dense polycrystalline salt may be at least an order of magnitude greater than velocities observed in

single crystal experiments. The investigators found that brine prefers to migrate along paths of high crystalline activity (i.e., grain and subgrain boundaries and crystal defects) and may be enhanced along these paths.

Except for the Salt Block and Biggers and Dayton (1982) studies, none of the laboratory studies discussed above was concerned with intercrystalline migration, and the Biggers and Dayton study was basically qualitative. Another uncertainty with the studies is the spatial effects. The majority of these studies dealt with small salt samples, usually single crystals. Spatial effects are better understood in the situ studies discussed below.

### In Situ Studies

There have been a limited number of brine migration tests which have been in situ, i.e., performed in salt mines. Studies were conducted at the Carey Salt Mine in Hutchinson, Kansas (Project Salt Vault), in the late 1960's and early 1970's (Bradshaw and McClain, 1971). Brine was observed to migrate toward a heat source and was collected as condensate. Substantially more moisture was collected in a room in which there was "considerable" shale.

A five-year field testing program which included brine migration tests was performed in the Avery Island, Louisiana, salt mine (Krause, 1983a, 1983b). Three experimental configurations were set up: (1) natural brine movement with no heating; (2) natural brine movement under elevated temperature conditions; and (3) synthetic brine movement under elevated temperature conditions. For a complete description of the experimental set-up, see Krause (1983b). Tests were run for approximately one year. The measured borehole wall temperature was approximately 51°C at heater mid-height and the calculated temperature gradient was approximately 1.2°C/cm in the immediate vicinity of the borehole wall. The effective moisture content of the salt was determined to be about 0.054 weight percent. The salt contains about 10% anhydrite. Approximately 23 grams of moisture were collected in the experiment using natural brine under elevated temperature and approximately 18 grams in the synthetic brine experiment (collection rates ranged from 0.026 to 0.044 grams/day) (Krause, 1983b). During the cooling-down period of the synthetic brine test, the rate of moisture collection increased significantly. Subtracting the brine collected in the unheated test gives a net accumulation of approximately 10 grams of moisture collected in the natural brine test and 5 grams in the synthetic brine test (before cooling).

The investigators found that the region of significant brine migration in these tests was very small, i.e., about 5 mm from the heater surface after one year. Tracers (deuterium water and magnesium) were observed to migrate toward the heat source (about 60 mm in an unspecified amount of time), although there had not been sufficient time for them to reach the heater borehole. The synthetic brine was not observed to migrate into salt crystals (nor was any salt dissolution observed), leading the investigators to conclude that the brine moved along microcracks or grain boundaries.

Brine migration tests are also being conducted in the Asse salt mine in West Germany (Westinghouse, 1983). The investigators plan to run tests for longer durations (up to two years) and higher temperatures (over 200°C) than were conducted at Avery Island. The investigators are also interested in testing the effects of radiation, stress, gas pressure, and petrology and mineralogy on brine migration. In addition to modeling salt types in West Germany, tests conducted in the Speisesalz (pure salt) are designed to provide information pertinent to salt domes in the United States. Initial results from Asse have shown that increasing pressure in the collection borehole decreases the amount of brine collected, suggesting that pressure gradients significantly affect brine migration (Rothfuchs et al., 1984).

#### V. MODELING OF BRINE MIGRATION

The first investigators to formulate a relatively complete mathematical model for predicting theoretical brine migration were Anthony and Cline (1971, 1972, 1974; Cline and Anthony, 1971). Their equation for all-liquid intracrystalline inclusion migration is:

$$v = D \frac{C_1}{C_s} \left[ \frac{1}{T} \cdot \frac{C_1}{C_1} + \sigma \frac{G_1}{RT} - \frac{1}{L} \frac{K}{L} + \frac{4\gamma V_s}{XL} \right] \quad (1)$$

where

- $v$  = droplet velocity
- $D$  = diffusion coefficient
- $C_1$  = concentration of salt in brine droplet
- $C_s$  = concentration of salt in solid
- $\sigma$  = Soret coefficient
- $G_1$  = temperature gradient in brine droplet
- $R$  = gas constant
- $T$  = absolute temperature, °K
- $K$  = kinetics potential at interface
- $L$  = dimension of droplet parallel to temperature gradient
- $X$  = dimension of droplet perpendicular to temperature gradient
- $\gamma$  = grain boundary tension
- $V_s$  = partial molar volume of solid

Equation 1 can be simplified conceptually as:

$$v = v_T + v_\sigma - v_K - v_\gamma \quad (1a)$$

where,

- $v_T$  = velocity due to ordinary diffusion
- $v_\sigma$  = " " " thermal diffusion
- $v_K$  = " " " kinetics at interface
- $v_\gamma$  = " " " surface tension at grain boundaries

Cheung and others (1980) evaluated the Anthony and Cline model and compared it with several earlier, more simplistic models. They found that the first two terms (i.e.,  $v_T$  and  $v_\sigma$ ) in equation 1a dominated the calculated velocity. The final two terms,  $v_K$  and  $v_\gamma$ , had relatively little effect on the calculated velocity. Furthermore, although the equation is the most complete available, it requires data for the final two terms that is presently difficult or impossible to measure. Roedder and Chou (1982) found that the estimation of diffusion coefficients by Cheung and others (1980) at elevated temperatures has no scientific basis and may not be conservative.

Researchers at the University of California-Berkeley have produced a large volume of work on the theoretical modeling of intracrystalline brine migration (Olander and Machiels, 1979; Olander and others, 1980; Pigford, 1982; Olander and others, 1982; Yagnik, 1983; Olander, 1984). Their work expanded on the work of Anthony and Cline by including the variables for kinetic and surface tension effects on migration velocities. For a complete discussion of their work, see Yagnik (1983) and Olander (1984). Some investigators (see Roedder and Chou, 1982; Chou, 1983) have disagreements with the Berkeley work, believing that the magnitude of interfacial kinetics and surface tension effects are overestimated and the existence of a threshold temperature is unproven (see section VI). Consequently, Chou (1983) believes that calculations made by the investigators at Berkeley underestimate the migration of fluid inclusions in salt, perhaps by several orders of magnitude.

Some empirical models have also been developed and, due to the lack of data for the theoretical models, are enjoying widespread use. Jenks developed the so-called Jenks equation, an empirical equation based on the work of Bradshaw and Sanchez (1968) on the Salt Vault Project (Jenks, 1972; 1979; Jenks and Claiborne, 1981). The curve-fit equation is based on migration rate versus temperature data:

$$\log V/G_s = 0.00656 T - 0.6036 \quad (2)$$

where  $V$  = migration velocity (cm/yr)  
 $G_s$  = temperature gradient ( $^{\circ}\text{C}/\text{cm}$ )  
 $T$  = temperature ( $^{\circ}\text{C}$ )

Jenks and Claiborne (1981) compared rates calculated by the Jenks equation with experimental data and concluded that the equation gives conservative values, although all the data do not fall below the curve. The equation is incorporated into a computer program, MIGRAIN, which was validated with data from the Salt Block II experiments (Hohlfelder, 1980). McCauley and Raines (1984) developed a modification of the MIGRAIN code, called BRINEMIG, which was also validated with Salt Block II data. BRINEMIG also uses the Jenks equation to model brine migration. Roedder and Chou (1982), however, disagree with the assertion by Jenks and Claiborne (1981) that the equation is conservative. Some of their concerns include: (1) that the assumed quantities of fluid in

the salt are too low; (2) that the Soret coefficient used is too low, by at least one half; (3) that the assumption of a threshold gradient for brine migration was not proven; (4) that assuming that salt formations are homogeneous is non-conservative because it ignores the effect of areas of anomalously high permeability; and (5) the assumption that brine migration can be modeled as if in single crystals is incorrect because it ignores intercrystalline water in the analyses. Roedder and Chou (1982) believe that these assumptions lead to in-flow calculations that are low, perhaps by several orders of magnitude.

## VI. UNCERTAINTIES IN PREDICTION

There are a number of uncertainties involved in the prediction of brine migration in a potential HLW repository in salt. Perhaps the most significant uncertainty involves intracrystalline versus intercrystalline inclusion behavior. Intracrystalline migration of brine inclusions is a fairly well understood process, but intercrystalline migration is not. Roedder (1984) suggests that intercrystalline fluids could comprise up to 50% of the total water present in a salt deposit. The volume and velocity of brine transported by intracrystalline migration may be insignificant relative to intercrystalline migration. Modeling of brine migration has generally considered only intracrystalline inclusions (see Jenks and Claiborne, 1981; McCauley and Raines, 1984), assuming that intercrystalline migration rates will not exceed intracrystalline migration rates. The conservatism of this assumption, however, is questionable. Intercrystalline migration may be driven by pressure gradients instead of temperature gradients, thus undermining the basis for the above assumption. Processes affecting the pressure in the salt, such as creep, may therefore be of considerable importance to brine migration. It is clear that the process of intercrystalline fluid migration needs to be studied more thoroughly before the assumption that intercrystalline migration is not greater than intracrystalline migration can be fully evaluated.

A related concern is the uncertainties regarding inclusion behavior at grain boundaries. When an intracrystalline inclusion reaches the grain boundary, various investigators observed that it either ceased migrating, crossed into the adjacent crystal, or began migrating along crystal boundaries. Its behavior depends on a number of parameters, including inclusion size (small inclusions were trapped), the nature of the boundary (i.e., how tight it is), and the magnitude of the temperature gradient. Since most of the fluids present in salt crystals are contained in the large inclusions, there is the potential that the majority of the fluids will not stop migrating at crystal boundaries under a temperature gradient. It is apparent that long term, in situ experiments need to be run in order to determine the behavior of fluids at grain boundaries.

Many salt deposits contain significant amounts of hydrated or hydrous minerals -- percentages greater than five are common with great local variability. Repository-induced conditions may cause these minerals to dehydrate, thus

producing fluids available for migration. However, it is unclear how these fluids will migrate, especially in clastic interbeds within the salt. The movement may be toward the surface, due to density gradients; thus, the impact will depend on the site stratigraphy to some extent. It may be incorrect to include dehydration fluids in intracrystalline migration models.

The existence of a threshold temperature gradient below which inclusion migration will not occur is a source of debate among investigators. Some investigators (see, e.g., Cline and Anthony, 1972; Jenks and Claiborne, 1981; Pigford, 1982; Yagnik, 1983; Olander, 1984) believe that a threshold gradient does exist in salt. Jenks and Claiborne (1981) calculated a series of values for the threshold gradient, while McCauley and Raines (1984) used a threshold gradient of  $0.125^{\circ}\text{C}/\text{cm}$  in their model, BRINEMIG. Other investigators, however, dispute the concept of a threshold gradient (see, e.g., Roedder and Chou, 1982; Chou, 1983). The disagreement centers around the magnitude of interfacial kinetics and grain boundary surface tension effects (Chou, 1983). Roedder and Chou (1982) also suggest that experimental data used to support the concept of a threshold gradient (Cline and Anthony, 1972) may not be applicable, and that other data (Roedder and Belkin, unpublished data; see Roedder and Chou, 1982) suggest that no threshold gradient exists. A scoping study by St. John and others (1982) suggest that a temperature gradient of  $4^{\circ}\text{C}$  over 300 meters will exist in a repository in salt at 1,000 years after waste emplacement, although a more recent study suggests that temperature gradients will be insignificant well before that (McCauley and Raines, 1984). The use of a threshold gradient by McCauley and Raines (1984) does not increase the predicted amount of brine accumulation at the waste package to a great extent (approximately 10% for civilian high-level waste). Because there is uncertainty about the existence of a threshold gradient, conservative predictions presently should not incorporate it. Additional experimental data modeling repository conditions should be collected to examine the threshold gradient concept.

The construction of a HLW repository and emplacement of waste may change the geochemical environment significantly enough to affect predicted brine migration rates and accumulated brine at the waste package. The actual construction may cause a number of changes, including fracturing of the salt, formation of brine inclusions, and the introduction of atmospheric oxygen. Fracturing will form pathways for fluid migration near the waste package; the extent of this problem will depend on the mining method used. Inclusions formed in the salt will probably be liquid-vapor inclusions. These inclusions should migrate down a temperature gradient, and their effect on the migration of all-liquid inclusions up the gradient is uncertain. Atmospheric pressures in the mined cavities will create a pressure gradient with respect to the salt rock that is under lithostatic pressure and possibly induce migration of intercrystalline fluids.

The irradiation of the rock salt by HLW may significantly influence the chemical environment. Based on Salt Vault data, Jenks (1979) suggested that irradiation will not affect brine migration, but more recent studies may contradict this. Radiation effects include the formation of colloidal sodium,

dramatic changes in the pH, and gas formation (predominantly H<sub>2</sub>; also CO<sub>2</sub>, water vapor, and possibly O<sub>2</sub>) (Panno and Soo, 1984). Radiation may also result in very high solution redox potential (1200 mv) (Gray and Simonson, 1985). Migrating brine may react with free chlorine or chlorine gas liberated during colloidal sodium formation to form HCl, a strong corrosive agent. The nature and magnitude of these effects depends on the moisture present near the waste package during irradiation, impurities in the salt, and the rate and duration of the irradiation.

Some investigators observed the decrepitation of fluid inclusions in salt at high temperatures in a short period of time (e.g., see Roedder and Belkin, 1979a). Decrepitation may cause extensive fracturing near the waste package and thus affect the nature and rate of brine migration. This breaking up of inclusions, however, was not always observed in brine migration experiments. The effects of decrepitation on brine migration are unknown.

## VII. SUMMARY AND CONCLUSIONS

The migration of brine inclusions in a salt deposit under a temperature and/or pressure gradient may affect the ability of a repository to isolate HLW from the accessible environment. Investigators have observed and modeled intracrystalline fluid behavior under a temperature gradient in a number of laboratory and some in situ experiments. This process is fairly well understood for both all-liquid and liquid-vapor inclusions. Intercrystalline fluid behavior, on the other hand, is poorly understood. The amount of fluids present in intercrystalline inclusions, however, can be significant. Fluid inclusion behavior under temperature and pressure gradients merits further study. Models predicting migration rates and amounts of fluid expected to reach the waste package in a HLW repository have incorporated only intracrystalline fluid behavior.

Additional uncertainties in predicting fluid behavior in a HLW repository in salt include behavior at grain boundaries, the migration behavior of fluids released by mineral dehydration, the possible existence of a threshold gradient, construction effects, radiation effects, the effects of redox conditions, and the occurrence of decrepitation of fluid inclusions. All of these uncertainties need to be better defined in order to accurately model brine migration in a HLW repository in salt.

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