Fluid Inclusions in Core Samples
from ERDA No. 9 Borehole,
WIPP Site, New Mexico
Status Report 2, Jan. 1, 1978
Edwin Roedder and H.E. Belkin

Introduction

Work on this project started on 20 Dec. 1976, with the receipt of 10 core samples. A preliminary status report on the results obtained during a reconnaissance scoping of the problem was issued 27 June 1977 (Roedder and Belkin, 1977), and the present report is the second in the series. A talk dealing with some of the data in the first report was presented at the 1977 Midwest AGU Meeting; the abstract has yet to be published in EOS. Information in the first report will not be repeated here, but the Interpretation sections here are based on the data in both reports. Relatively little new information is ready for reporting at this time, in part due to delays in arranging funding.

Samples studied

In addition to continuing studies on the samples obtained previously, nine new samples from ERDA No. 9 were selected on a visit to Carlsbad and processing started. These are from the following footage intervals, and were selected to concentrate work on the horizons of maximum interest:
2058.8 - 2059.0
2070.4 - 2070.6
2606.5 - 2606.9
2617.2 - 2617.7
2626.7 - 2627.0
2659.0 - 2659.2
2665.0 - 2665.1
2692.4 - 2692.6
2699.8 - 2700.0

In addition to the above "representative" samples, four nonrepresentative samples from ERDA no. 9 were selected because they contained plainly visible large inclusions, suitable for special tests:

2061.2 - 2061.6
2064.5 - 2065.0
2614.7 - 2615.0
2618.5 - 2619.0

In order to test certain sample preparation and inclusion extraction procedures with no loss of important core, two pieces of core from the AEC 8 core were also picked:

2059.5 - 2059.8
2462.0 - 2462.9

In the Kermac Mine, Lea County, New Mexico, northwest of the WIPP site a 4-m igneous dike has cut potash ore beds. As this represents a natural simulation of some of the heat effects of canister storage, we thought it would be instructive to examine the fluid inclusions in
samples taken at measured distances from this dike. The samples, collected and kindly provided by Prof. M.W. Bodine, Jr., of the New Mexico Institute of Mining and Technology, have the following identifications:

- MB-77-8  White halite 1-2 cm from dike
- MB-76-3  Barren salt 0.2 m from dike
- MB-76-4  "Ore" horizon 2.5 m from dike
- MB-76-5  "Ore" -21 m from dike

Of these samples, the field evidence indicates that only MB-77-8 has been molten. The others show some mineralogical effects of the heating, but as sedimentary structures are preserved, they have presumably not melted (personal communication, M. Bodine).

**Sample preparation**

Essentially as reported earlier.

**Methods of study - decrepitation**

In addition to the methods reported earlier, some decrepitation tests were run. In these a portion of the core weighing approximately 100 g was split out with a rock splitter; where possible the outside core surface was avoided, due to the probability of physical deformation and resultant leakage of inclusions. This piece was wrapped loosely in \(\{\text{aluminum foil}\}\), weighed, and heated to the run temperature over \(\approx 8\) hours to avoid thermal shock, held at run temperature for 3-4 days, cooled and reweighed to determine weight loss. Following this the homogenization
temperature range was redetermined for a series of inclusions that appeared, on petrographic examination, to be representative. Crushing tests were also repeated on the heated samples to detect the presence of noncondensable gases under pressure.

Results

Petrographic examination.

Although the nine new samples from ERDA no. 9 have been examined in detail, inclusion counts have not yet been made. However, we have no reason to doubt that they will be similar in range and amount of inclusions to those found in the first suite of samples. Most smaller inclusions have no bubbles at all, and the larger inclusions generally have bubbles estimated to homogenize in the same range as before (i.e., 20-45°C).

Decrepitation tests.

These tests were run only on the nine new samples. A summary of the test data and results is given in Table 1. On examination of the material after the decrepitation tests, several general features were evident. The nature of the test is such that systematic before and after inclusion counts on the same sample are impossible, but it was obvious that although most of the larger inclusions had decrepitated, and were empty, many small inclusions (≤ 100 μm) had not. These small inclusions appear perfectly normal in distribution, but almost all now have appreciable vapor bubbles (Fig. 1), with a vapor/liquid ratio that is obviously higher than that present in two-phase inclusions before
Table 1. Test data and weight loss (in %) from decrepitation tests.

Roman numerals indicate amount of sample breakup after run:
I - unbroken; II - broken into 2 or 3 pieces; III - broken into more than 3 pieces.

<table>
<thead>
<tr>
<th>Temperature, °C</th>
<th>Core interval (ft.)</th>
<th>150</th>
<th>200</th>
<th>250</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2058.8 - 2059.0</td>
<td>0.16 II</td>
<td>0.23 II</td>
<td>0.76 III</td>
</tr>
<tr>
<td></td>
<td>2070.4 - 2070.6</td>
<td>0.16 II</td>
<td>0.12 I</td>
<td>0.35 III</td>
</tr>
<tr>
<td></td>
<td>2606.5 - 2606.9</td>
<td>0.02 I</td>
<td>0.24 II</td>
<td>0.95 III</td>
</tr>
<tr>
<td></td>
<td>2617.2 - 2617.7</td>
<td>0.06 I</td>
<td>0.30 I</td>
<td>0.62 II</td>
</tr>
<tr>
<td></td>
<td>2626.7 - 2627.0</td>
<td>0.19 I</td>
<td>0.28 I</td>
<td>1.04 III</td>
</tr>
<tr>
<td></td>
<td>2659.0 - 2659.2</td>
<td>0.14 I</td>
<td>0.13 I</td>
<td>0.59 III</td>
</tr>
<tr>
<td></td>
<td>2665.0 - 2665.1</td>
<td>0.19 I</td>
<td>0.18 I</td>
<td>0.75 III</td>
</tr>
<tr>
<td></td>
<td>2692.4 - 2692.6</td>
<td>0.06 I</td>
<td>0.26 I</td>
<td>0.83 III</td>
</tr>
<tr>
<td></td>
<td>2699.8 - 2700.0</td>
<td>0.19 I</td>
<td>0.24 II</td>
<td>0.65 III</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td>0.13</td>
<td>0.22</td>
<td>0.73</td>
</tr>
</tbody>
</table>

Heating schedule (hours)

<table>
<thead>
<tr>
<th></th>
<th>Room T to run T</th>
<th>7</th>
<th>9.5</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hold at run T</td>
<td>79</td>
<td>95</td>
<td>79</td>
<td></td>
</tr>
<tr>
<td>Run T to room T</td>
<td>~9</td>
<td>~9</td>
<td>~10</td>
<td></td>
</tr>
</tbody>
</table>
heating. Only the 150°C sample still had inclusions without bubbles; a few 10-20 μm one-phase liquid inclusions were found there. All three samples showed some "steam" inclusions—cubic negative crystals with fillets of liquid in the corners. No gas under pressure was found in any of the heated inclusions. In most cases, the weight losses increase with increasing run temperature; the several exceptions are probably a result of nonhomogeneous distribution of inclusions in the sample aliquots taken.

Homogenization temperatures were determined on two-phase inclusions remaining in the samples after the decrepitation tests. The results are given in Table 2, along with the volume percent of vapor phase, estimated from measurements of bubble diameters.

Table 2. Temperature of homogenization and volume % vapor of two-phase inclusions in salt of samples after decrepitation tests.

<table>
<thead>
<tr>
<th>Decrepitation run temp. (°C)</th>
<th>Homogenization temp. (°C)</th>
<th>Number of inclusions</th>
<th>Volume % vapor phase; avg. for ~25 inclusions</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Minimum</td>
<td>Maximum</td>
<td>run</td>
</tr>
<tr>
<td>150</td>
<td>90</td>
<td>120</td>
<td>16</td>
</tr>
<tr>
<td>200</td>
<td>110</td>
<td>180</td>
<td>21</td>
</tr>
<tr>
<td>250</td>
<td>180</td>
<td>273</td>
<td>28</td>
</tr>
</tbody>
</table>

Note: Birefringent crystals, present in some of these inclusions, do not disappear during homogenization runs.
Study of suite of samples from the Kermac mine

The three samples closest to the dike yielded usable inclusions. (Sample MB-76-5 was too finegrained and opaque to be usable by the sample preparation techniques used here, and will not be considered further in this report.) In all three samples about 15% of the small inclusions (< ~20 \( \mu \text{m} \)) were without bubbles as received. Most inclusions in all three samples were normal, two-phase inclusions with a small bubble. Homogenization temperatures were determined on a representative group of these two-phase inclusions in each, with results as shown in Table 3.

Table 3. Temperature of homogenization of two-phase inclusions in salt from near dike in Kermac mine, Lea County, New Mexico.

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Distance from dike (m)</th>
<th>Temp. of homogenization (°C)</th>
<th>Number of inclusions run</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Minimum</td>
<td>Maximum</td>
</tr>
<tr>
<td>MB-77-8</td>
<td>0.01 - 0.02</td>
<td>71</td>
<td>116</td>
</tr>
<tr>
<td>MB-76-3</td>
<td>0.2</td>
<td>60</td>
<td>110</td>
</tr>
<tr>
<td>MB-76-4</td>
<td>2.5</td>
<td>85</td>
<td>88*</td>
</tr>
</tbody>
</table>

* Two birefringent crystals remained unchanged, and a cubic crystal (KCl ?) dissolved at 82°C. (See Fig. 2 for a similar inclusion.)
Freezing runs were made on inclusions in two of these samples. In both cases the first melting temperatures were well below that of a pure NaCl-H₂O system (≤-28°C for MB-77-8 and ≤-31.0°C for MB-76-4). On warming, the last solid phase to dissolve in both samples was an unidentified phase, presumably a hydrate other than NaCl·2H₂O. This dissolved at +11 to +18°C (MB-77-8) and at +4 to +7.2°C (MB-76-4).

One 75-μm inclusion was found in MB-77-8 that yielded an entirely different freezing behavior. This inclusion contains a single homogeneous fluid at room temperature, presumed to be gas (Fig. 3a). On cooling to the range -70 - 75°C this inclusion develops a number of grains of an unidentified solid (Fig. 3f). On warming to about -68°C, a liquid/vapor meniscus becomes visible (Fig. 3e), with solid grains still present. The last of these grains disappears at ~-56°C (Fig. 3d); further warming on a the liquid/gas meniscus becomes faint (Fig. 3c) and the two homogenize, by expansion of the liquid, at -20.9°C (Fig. 3b). This behavior is difficult to interpret in terms of composition. CO₂ has its triple point at -56°C, in exact agreement with the disappearance of the solid phase at -56°C observed here, but obviously the inclusion cannot contain just CO₂, as there is liquid present at a lower temperature. The nature of the mixture of gases can only be guessed at this time.

The crushing tests on the Kermac samples were most revealing. The normal, small-bubble inclusions contain a vacuum bubble (i.e., water vapor at ~20 mm pressure). Many inclusions that either had a very large bubble or appeared to contain gas only, from all three samples, were found to contain either a vacuum (Figs. 4, 5), or a partial vacuum
(Figs. 6, 7). These are probably from the trapping of bubbles of steam, with or without some noncondensable gas and brine. All three samples also contained a few inclusions with gas under greater than atmospheric pressure. These inclusions were one-phase as first observed, and except under special circumstances, it is not possible to distinguish between a vacuum, dense gas, or even liquid, in such single inclusions. It is only on crushing, when these formed bubbles of noncondensable gas in the surrounding fluid, that the internal pressure became evident. By measuring the size of the inclusion before crushing, and the diameter(s) of the bubbles evolved, we obtain a crude measure of the internal pressure, assuming each one volume expansion corresponds to one bar pressure. In a few inclusions, the contents change to a two-phase, liquid + vapor system during the expansion (Fig. 8), but usually the pressure release is too sudden to tell if there was a transient two-phase condition. The volume expansion varies for different inclusions, from 30- to 40-fold (Figs. 9 and 10) to a maximum of perhaps 100-fold (Fig. 11). As the immersion medium used is an oil in which methane is probably readily soluble, this gas is more likely CO₂, since the bubbles dissolve very slowly.

Interpretation of the data from the Kermac mine samples is difficult at this time. The presence of an inclusion filled with a dense CO₂-rich (?) gas in sample MB-77-8, that was probably molten at the time of intrusion of the dike, is not unexpected, since there is a small amount (0.1-2%) of magnesite in the adjoining rock (M. Bodine, personal communication). The dike shows no evidence of carbonate alteration,
however, and there is a problem in explaining the presence of inclusions of low-temperature, gas-free brine, apparently pure steam, and dense CO₂ in the same sample. Formation of these types at different times is obviously required, but there is no other indication of differences in conditions or time of origin.

The temperature at which the dense gas inclusion was trapped is of interest. Although pure NaCl melts at 800°C, other materials present here (such as KCl) will drop this temperature. If we assume that the gas inclusion contains pure CO₂, the -21°C homogenization indicates a filling density of about 1.03 g·cm⁻³ (Roedder, 1965). Combining this with the estimate of confining pressure of ~38 MPa (380 bars) yields an obviously erroneous "trapping temperature" of 0°C. The several inconsistencies and difficulties in interpretation of the data from the Kermac samples cannot be resolved until additional studies are made.
Interpretation - Geological significance of inclusion data*

The fluids from which the salt crystals making up these beds originally crystallized were exceedingly saline brines with much material other than NaCl in solution, as evidenced by the freezing data on primary inclusions in hopper salt. These hopper crystals appear to have grown on the surface and then have sunk, as described by Dellwig (1955). All but about 1% of this original hopper salt texture has been eliminated by recrystallization at some unknown later time, yielding coarser, clearer crystals of salt, and presumably gross changes in the mineral assemblage other than halite. Some of the clear salt surrounding the chevron structures may represent crystallization on these hopper nuclei on the bottom of the basin, but most is believed to be from recrystallization. This recrystallization occurred in the presence of exceedingly saline brines, also with much material other than NaCl in solution; this fluid may have been essentially fluid from the primary fluid inclusions, released during the recrystallization. It has been trapped as large inclusions in the recrystallized salt and comprises the bulk of the liquid now present in the samples as studied in the laboratory. Although the salt beds now appear dry to the eye, the samples contain 0.1 to 1.7 weight % fluid as examined, and may contain double these amounts in situ. At some stage or stages in the history of these beds, the fluids present in these rocks varied in composition. A very few were saturated with organic gases such as methane and

*Based on data from both this and the previous report (Roedder and Belkin, 1977).
actually contained bubbles of a separate gas phase. Others contained additional salts in solution, resulting in the precipitation of various crystalline solids from the liquid within inclusions in halite. The relative time sequence of these various fluids is not known, and most inclusions have neither methane nor crystalline solids.

Homogenization temperatures of inclusions in recrystallized salt range from 20.4 to 45.5°C. During burial, creep from recrystallization in a salt bed with intergranular films of solution should result in the hydrostatic pressure on fluids during trapping in a crystal being essentially equal to the lithostatic pressure. If an overburden of 5000 ft. (1524 m) is assumed to have been present during the recrystallization, the maximum pressure (lithostatic) would have been >380 MPa (380 bars). There are no PVT data on the specific fluids present in the inclusions, but if the data on a 25-percent NaCl solution are used, this pressure would suggest a maximum pressure correction of 36°C (Potter, 1977), and hence trapping (i.e., recrystallization) temperatures of 56-82°C. However, the true pressure correction may be lower, particularly since the fluids are more saline than 25-percent NaCl. Thus there is a drop of 12°C in the pressure correction on changing from 20-percent NaCl to 25-percent NaCl.

The presence of smooth planes of primary hopper salt inclusions of various sizes, separated by planes of inclusion-free salt, with the plane orientation horizontal, proves that these minute inclusions have not moved measurably since deposition, although they have been in a geothermal gradient presumably similar to that of the present for about
225 m.y.

The widespread occurrence of metastable phenomena in these inclusions, even in some large ones, suggests that fluid flow in the past through these beds has been very slow. Also, the fact that all inclusions that have been frozen prove to contain bitterns, and none was found with just NaCl-H₂O solution, proves that at no time during the history of these samples did ground water, saturated only with respect to NaCl, ever penetrate into these particular samples.

**Interpretation - Significance of inclusion data to the design of an atomic waste repository.**

Amount of water in the beds

Although this is the most important single datum that might be obtained from inclusion studies, we can only provide minimum values. The nature of the samples and the necessary sample preparation are such that there is a bias toward low values built in to these data. Thus the 0.1 to 1.7 weight percent fluid actually found in these individual samples may represent perhaps only half (or even less) of the liquid that was present in these beds in the ground before sampling. Superimposed on this uncertainty in accuracy is an exceedingly poor precision in the measurement, due to gross variability of the sample material. Thus adjacent slabs of the same sample showed a five-fold range. Processing of much larger samples should help to eliminate much of this

*Based on data from both this report and the previous report (Roedder and Belkin, 1977).
variation. It is possible that neutron logs, reflecting the amount of hydrogen in the beds, might give a more accurate evaluation of the total water present, since they would effectively integrate over a much larger sample and would include the effects of large liquid-filled intergranular cavities which may be missed.

One important aspect of the inclusions on the properties of the salt is the effect of the constituents present on the lowering of the temperature of melting, and on the amount of liquid present at any given temperature. In this connection it is important to note that the inclusion fluid is not water, but a strong bittern with probably significant quantities of calcium ions. This will certainly affect the temperature coefficient of solubility of NaCl in these liquids, and will have important effects on the vapor pressure of these liquids (see beyond).

Corrosive gases in the inclusions.

Although gas analyses by mass spectrometry are still to be made on the inclusion fluids, it is evident from the crushing studies that the inclusions in these samples, at least, contain very little noncondensible gases. Although hydrogen sulfide was found as an important constituent of gases in a gas pocket in another drillhole, there can be very little, if any, in these inclusions. The human nose is a rather sensitive detector of H$_2$S, ordinarily responding to a minimum of $\sim 10^{-10}$ g. No odor of H$_2$S was detected during sample preparation, when relatively large volumes of inclusion liquid were exposed to the air. Gases such
as HCl formed within the inclusions or by reactions of inclusion fluids with other minerals during heating, and under intense gamma radiation, may be a much more important cause for corrosion than those now present in the inclusions. Water itself, however, would be an effective corrosion agent for hot steel components, by conversion to iron oxide and hydrogen.

Effects of water on physical properties of the salt

The intergranular porosity in these salt beds, in situ, is probably saturated with solution; such fluid films are probably responsible for at least some of the flowage phenomena in natural salt, and may make for large differences between the physical properties, and behavior, of natural salt beds vs those shown by essentially dry samples in the laboratory. Although it may be difficult to set up valid experiments to quantify the effects, the presence of such solutions certainly will have gross effects on the physical strength and the rates of deformation.

Such fluids will tend to be squeezed out of the salt beds over geological time as recrystallization along grain boundaries and other processes permit compaction, just as the connate water in a sandstone is eliminated during the formation of a quartzite. Such flow will occur, however, only if there is an escape route for the fluid to take; as pointed out very aptly by Baar (1977), there is much evidence from high pressure fluid pockets that salt rocks in situ are generally impermeable at depths exceeding about 300 m. It may well be that under
such conditions, that preclude further "dewatering", the fluid in these intergranular films is still able to migrate short distances, enough to permit it to coalesce into "pockets", perhaps at points of slightly reduced pressure, just as a plastic shale will flow to the crest of folds in firmer rocks.

Even if the intergranular liquid were to be removed completely, yielding a polycrystalline texture with salt crystals in true contact with each other, there is no driving force to remove the fluid inclusions from within the individual halite crystals. If such a salt bed is then deformed to the point of rupture of salt crystals, any inclusions along the rupture will provide water to "lubricate" the fracture and hence lower the apparent strength of the rock. Although one might expect, intuitively, that a new fracture would preferentially follow planes of inclusions, as from the healing of former fractures, Gerlach and Heller (1966) have shown that the salt recrystallized in the vicinity of such a healed fracture is actually stronger than the adjacent unrecrystallized salt, and new fractures could not be made through inclusions there. Actually, most of the fluid in inclusions in the Carlsbad salt is in large, randomly arrayed inclusions in completely recrystallized salt, and examination of fractures through such samples suggests that propagating cracks have preferentially taken routes through such inclusions. Thus a mine pillar that might be thought to have had ample exposure to one atmosphere to permit thorough dewatering, could be expected, on deformation and release of inclusion fluids, to have lower strength than a truly dry salt pillar.
One example of the surprising effects of inclusion fluids on the properties of salt crystals is found in the behavior of fluid inclusions in single crystals of salt under uniaxial stress (Roedder and Belkin, 1977, p. 13 and Fig. 28). Under these conditions, fluid inclusions visibly change shape, and volume, within a few minutes, at room temperature. This is a new inclusion phenomenon, never reported before, but physically understandable; it may well be of consequence in the propagation of cracks through stressed, inclusion-bearing salt.

Evidence of previous solution movement in these beds.

The fluid inclusion evidence obtained so far on solution movements is encouraging in that it indicates at most only very slow movement, and suggests that part of the fluids now present in these beds may even be Permian in age. The lack of any pure NaCl-H₂O inclusions also argues against any previous sudden inrush of groundwater that could be trapped as inclusions in halite before it had time to acquire a full complement of other salts. Note, however, that this applies only to the specific samples examined. For example, the waters responsible for the formation of the solution cavities (breccia pipes) found in the Delaware Basin (Vine, 1976), probably were low in salinity, if this was the actual cause of the pipe formation. If low salinity water, whatever its origin, comes in contact with the bottom of a saline formation, an exceedingly effective and possibly rapid upward boring mechanism would result, driven by density currents. It would be desirable to examine secondary inclusions in material from these pipes.
Movement of inclusions in a thermal gradient.

All fluid inclusions should move when the host crystal is placed in a thermal gradient. The nature and rate of such movement has been the subject of numerous studies, since it is of consequence in the chemical industry. Wilcox (1968) summarizes this extensive work (111 references). Similar migration of inclusions is also an important cause for degradation of laser crystals grown at high temperatures by the Czochralski technique (Hopkins et al., 1976). Most liquid inclusions move up the thermal gradient, but if the vapor bubble is large relative to the liquid, and particularly if boiling occurs, the movement may be in the reverse direction (Wilcox, 1969; Anthony and Cline, 1972; Chen and Wilcox, 1972). The rate of movement is independent of inclusion size in many systems, but strongly (and directly) dependent on inclusion size in others (Wilcox, 1968); there may be a threshold size below which no movement occurs (≈10 μm; Anthony and Cline, 1971a). Large inclusions may break up during movement (Wilcox, 1968; Anthony and Cline, 1973). Many factors may affect the rate of migration, even in a given host, including gravity, composition, surface tension, inclusion shape, crystal anisotropy and imperfections, presence of a foreign gas, etc. In salt, since the thermal coefficient of solubility is small at surface temperatures, but increases greatly with increase in temperature, the rate of movement in a given gradient can be expected to increase with increase in ambient temperature. Higher ambient temperature also increases the travel rate by increasing the solubility, the diffusion coefficient, and the interface kinetics, and the increase in
rate was found to be particularly striking in NaCl (Wilcox, 1968, p. 20). The fact that the inclusions in Carlsbad salt have not moved measurably during geological time in the geothermal gradient is perhaps an indication of an exceedingly slow rate, or perhaps a result of other weak factors, such as gravity, counteracting the effect.

Accurate prediction of the behavior of in situ inclusions in the thermal gradient around a canister in salt cannot be obtained at this time, as there are too many uncontrolled variables. Since most inclusions at Carlsbad contain no bubble or only a very small bubble, they will move toward the canister, as was shown to occur at Project Salt Vault (Holdoway, 1974). The rate of this movement will depend, most particularly, on the inclusion size, the ambient temperature of the grain, the temperature gradient, and the temperature coefficient of solubility of NaCl in the particular inclusion fluid involved. The average distance that an inclusion must travel before it intersects a grain boundary will vary with the grain size of the salt, but seldom would be over 1 cm.

What can be expected when the inclusion reaches a grain boundary? Anthony and Cline (1971b) showed that grain boundaries tend to trap migrating droplets, but Anthony and Sigsbee (1971) showed that gas bubbles (10–50 μm) migrating in polycrystalline camphor could cross grain boundaries. However, migrating bubbles were observed to drag grain boundaries when the bubble concentration on the grain boundary was large. Although at first glance this is seemingly a very different process, it may actually present us with a fairly good model of the
salt-liquid system. In Project Salt Vault, Holdoway (1974) says only that "little migration across grain boundaries appears to have occurred."
Obviously, if a grain boundary is very tight, a large inclusion could cross it without effect. But if the grain boundary is at all open, the fluid could spread out along it as a film. Any such film should act as an inclusion, with material diffusing across it away from the heat source, and a net boundary movement toward the heat source. Such a large planar "inclusion" should break up to form new small inclusions in the next grain.

The behavior of an inclusion migrating to the wall of an open cavity (e.g., that holding the canister) will be different. On contacting the cavity, Anthony and Cline (1974) have shown that some evaporation occurs, a vapor bubble forms, the inclusion seals itself, and now that it has a large bubble, it reverses direction and moves back the way it came, this time going down the gradient.
Decrepitation release of liquid inclusions

In view of the expected rather slow migration rates of liquid inclusions in the thermal gradients established by the cannisters, it seems probable that release of inclusion fluid by decrepitation will be a much more important process. The effects to be expected are not as simple as they might first seem. An individual inclusion, if it is assumed to have rigid walls, to contain 25-percent NaCl solution, and to homogenize at 40°C, will develop 70.0 MPa (700 bars) internal pressure when heated to 100°C, and 200 MPa (2000 bars) at 200°C (Potter, 1977). However, salt cannot stand such internal pressures without yielding. Even a strong, hard mineral such as quartz starts to decrepitate when the internal pressure in its fluid inclusions reaches 80 ± 3 MPa (800 ± 3 bars) (Khetchikov and Samoilovich, 1970), and halite can be expected to decrepitate at far lower pressures, perhaps even an order of magnitude lower.

When Carlsbad halites were heated, some of the inclusions do decrepitate, in part causing the samples to crumble, and yielding an average weight loss of 0.13% (150°C), 0.22% (200°C), and 0.73% (250°C). This does not mean that just this amount of inclusions was opened; it means only that this much water (and other volatiles) left the system. Considerably more inclusion fluid may well have been exposed, and merely lost water until its concentration of salts was such that the vapor pressure of H₂O at that temperature was less than one atmosphere. It is important to remember that the salts in solutions in these brines (including materials other than NaCl as well) have
large effects on the vapor pressure and hence on any vapor transport.

In addition to those inclusions that decrepitated, however, many inclusions remain sealed as liquid inclusions in the sample after heating, albeit in a changed form. These have permanently deformed their walls to form a larger chamber, and now have a much larger shrinkage bubble, proportional to the amount of expansion. (Why some decrepitate and others expand is at present unanswered; residual stresses in the host crystal may be involved.) This expansion phenomenon is not new - it has been reported at various times in the past. It is the net result of several volume changes during heating of the inclusion. On heating, the host salt expands, enlarging the cavity as well. This effect is small, yielding a cavity volume increase of only 1.2% from 40 to 140°C. The fluid in the inclusion dissolves more salt from the walls on heating; the net volume effect; i.e., an increase in cavity volume) of this (generally a decrease will vary widely with the composition of the solution. Thermal expansion of the brine would develop high pressures if the walls were rigid, but instead, in the presence of liquid, the inclusion walls frequently just expand. The amount of expansion is roughly proportional to the temperature of heating, i.e., the inclusion fluid stretches the walls until its internal pressure drops just below that needed to cause further stretching at that temperature. Nucleation of a vapor phase may even occur (Geguzin and Dzyuba, 1973), in which case loss of water to the expanding vapor phase also helps to limit the expansion.
Some of the thermodynamic variables of this process have been modeled by Cline and Anthony (1971) for the pure salt system, but unfortunately there are several additional variables that make their modeling not directly applicable to the natural system. One of these, of course, is the more complex composition of the natural fluids, and another is the existence of a confining pressure on some of the salt around the cannister. Salt immediately adjacent to the cannister opening can be expected to decrepitate into the cavity on heating. But once a "back pressure" is built up by the apparent volume expansion to form decrepitated salt, further inward expansion will become increasingly difficult. Additional heat flow into the surrounding rock would normally cause simple thermal expansion of the rock itself, and still greater expansion from the inclusions. If there are significant confining pressures, both inclusion decrepitation and expansion will be retarded, since both are responses to differential stresses. The net result of these three sources of potential expansion should be examined carefully in light of the large-scale mechanical properties of rock salt beds, possible regional stresses that might be present, etc.

If the pressure buildup due to this expansion is slow enough to permit flowage of salt, it is possible that at least some of it might be accomodated by flowage closing up intergranular porosity and forcing out the fluid originally filling these pores.

**Future work**

We will use the techniques developed and the data obtained on
the original group of samples, as reported in the first report, to study the new samples. We also will extend some of these studies in new directions. In particular, we intend to perform chemical analyses of the gases in the inclusions, and of the ions present in solution, as well as obtain isotopic data on the water, to aid in understanding the origin of these fluids, and to permit better physical and chemical modeling of the expected behavior of these systems in an actual waste isolation system setting.
References


Figures. (In each, the length of the scale bar is 100 µm.)

1. Group of small primary hopper-growth inclusions in ERDA-9 sample 2699.8-2700.0, after 250°C decrepitation run. These inclusions probably had no bubble originally, and now have one as a result of plastic deformation of the host salt. They now homogenize at temperatures as high as 273°C (Table 2).

2. Solid phases (daughter crystals?) in inclusion in Kermac sample MB-76-4. The small rod-like crystal has parallel extinction, and the large cubic(?) crystal (KCl?) appears isotopic.

3. Dense gas inclusion in Kermac sample MB-77-8, photographed at the approximate temperatures indicated (°C). See text.

4, 5 - Steam inclusions (arrows), in Kermac samples MB-76-3 (Fig. 4) and MB-77-8 (Fig. 5), now containing essentially vacuum, before (a) and after being intersected by a fracture during crushing tests (b). The surrounding oil has filled the inclusions completely in (b).

6, 7 - Gas inclusions in Kermac sample MB-77-8 containing gas at less than one atmosphere pressure, before (a) and after being intersected by a fracture during crushing tests (b).

8. High pressure gas inclusion in Kermac sample MB-76-4 before (a) and after being intersected by a fracture during crushing (b).

9, 10, 11 - High pressure gas inclusions in Kermac samples MB-77-8 (Figs. 9 and 11) and MB-76-3 (Fig. 10), before crushing (a) and after being intersected by a fracture during crushing (b). The approximate volume expansion is 30-fold in Fig. 9, 40-fold in Fig. 10, and 100-fold in Fig. 11. Two bubbles formed in 9, and one in 10 (arrows).