Behavior of Fluid Inclusions in Salt During Heating and Irradiation

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ABSTRACT

Rock salt was heated and irradiated in situ by implanted radioactive wastes during the Project Salt Vault experiment, which was carried out at Lyons, Kansas, in the abandoned Carey Salt mine between 1965 and 1967. Petrographic examination showed that irradiation resulted in coloration of the salt, producing colors ranging from blue-black nearest the radiation source, to pale blue and purple farther from the source. Bleached areas are common in the radiation-colored salt, many representing trails produced by the migration of fluid inclusions towards the heat source. During laboratory radiation studies, it was found that these bleached areas frequently colored much less readily than ordinary colorless salt which had not been irradiated previously. This observation, and the relationship between primary chevron structures and migrated fluid inclusions, suggests that the bleached trails represent the total amount of migration of the inclusions from the beginning of Project Salt Vault, until the ambient temperature was reached after the conclusion of the experiment.

INTRODUCTION

An important factor to consider before the storage of radioactive wastes in salt can be initiated is the degree to which heating and irradiation will cause major changes in crystal structure and petrofabric of the salt. The main objectives of this investigation were to determine the general nature of the radiation damage caused by the gamma-ray component of the high energy radiation in salt, and to study water migration within the gamma irradiated salt.

The salt examined was irradiated in situ during the Project Salt Vault experiment. This experiment, sponsored by the Atomic Energy Commission, was carried out primarily by personnel from Oak Ridge National Laboratory, between November, 1965 and June, 1967 in the abandoned Carey Salt mine at Lyons, Kansas (Fig. 1). The salt beds in this mine belong to the Hutchinson Salt Member of the Wellington Formation, Leonardian Stage, Lower Permian. This member is composed of salt which is 87.5 percent pure and consists of units of interbedded salt, shale and anhydrite, and units of relatively pure salt. The section mined for the experiment is in relatively pure salt, with minor amounts of anhydrite.

Project Salt Vault was a demonstration of a method for the disposal of high level radioactive wastes. The aims of the experiment were to demonstrate waste handling equipment, and to determine the effects of radiation up to a dose

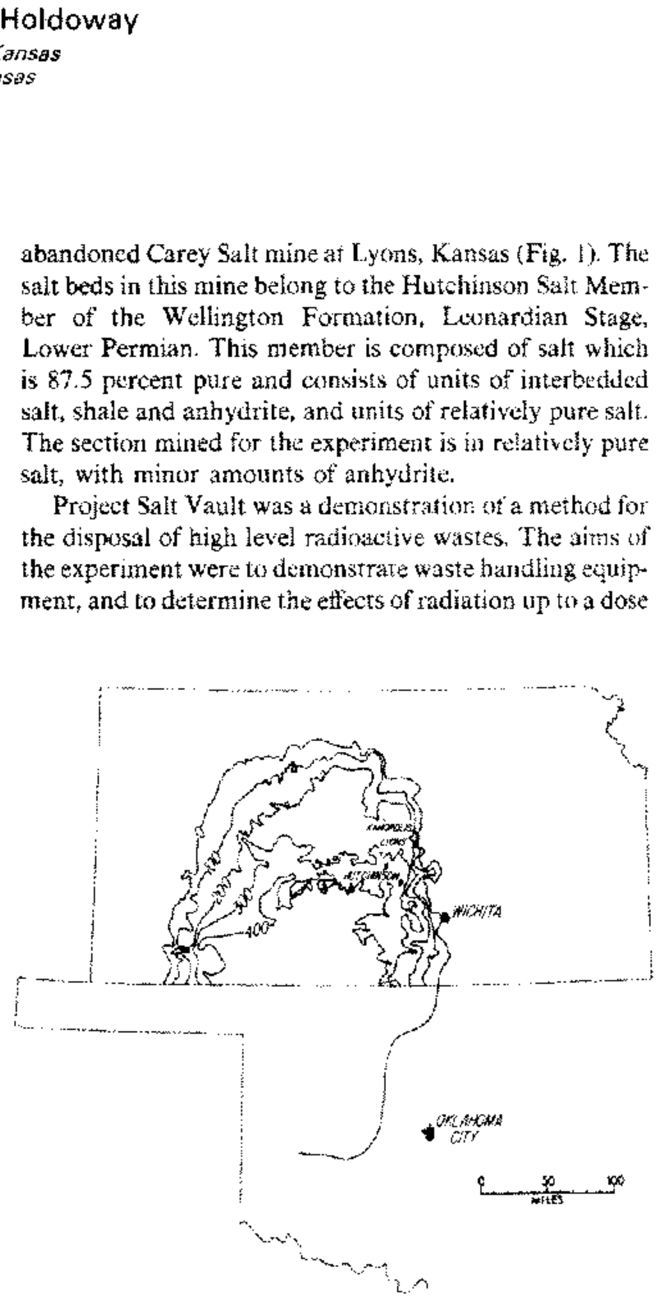

Figure 1. Map of Kansas showing area and thickness of the Hutchinson Salt Member of the Wellington Formation.

of 10^9 rads in salt at temperatures between 100 and 200°C. Preparation in the mine included the excavation of four rooms, and the drilling of seven holes in the floors of three rooms (Fig. 2). Of these three rooms, one was used for the main radioactive array, another for an electrical heating experiment, and the third for a subsidiary radiation study. A can containing two Engineering Test Reactor fuel assemblies was placed in each of the holes of the main array. In order to increase the radiation dosage received by the salt, the assemblies were exchanged every six months for freshly irradiated assemblies. Further details of the design and preparation for the demonstration were presented at previous symposia (Empson, et al., 1966, 1969).

During the Project Salt Vault experiment it was found that more water entered the array holes than had been anticipated. This water is thought to have been derived from fluid inclusions within the salt which migrated toward the radiation sources. If salt beds are to be used as a repository for radioactive wastes, such migration presents a possible hazard. On entering the array holes water would become contaminated with dissolved radioactive waste. Further migration of this water could lead to contamination of the water supply. One of the main objectives of the study was to try to determine to what extent migration of fluid inclusions occurred during Project Salt Vault, and how the migration was affected by the radiation field.

Oriented cores of salt irradiated in situ during the Project Salt Vault experiment were cut in August 1971. The cores were cut at an angle of 45° on the outside of two holes of the main array, entering the holes 2.7 m below the mine floor. Each core is 10 cm in diameter and has been cut into quarters lengthwise (Fig. 3). Quarter B from hole 2 was used in this study. The core was studied from its intersection with the array hole to a point 90 cm along its length. This corresponds to 0 to 65 cm from the array hole. Radiation-induced color is seen between 0 and 63 cm along the core (0–45 cm perpendicular to the array hole). The main part of the study was concerned with petrographic examination of thin sections. In addition, small pieces of irradiated salt were studied under a scanning electron microscope, and a series of thermoluminescence glow curves was run on both irradiated and unirradiated salt. Before thin sections were prepared, the core was embedded in bioplastic to which a small amount of red dye had been added to aid its recognition in thin section. Slices were cut from the faces of the quarter of the core, mounted on micro slides, and ground dry with sandpaper. Low magnification photographs were taken of thin sections before they were studied in detail under the microscope, in case transmitted light caused any bleaching of the radiation-induced color. Later, thermoluminescence studies showed that a temperature of at least 300°C is necessary for decoloration. It can therefore be assumed that no

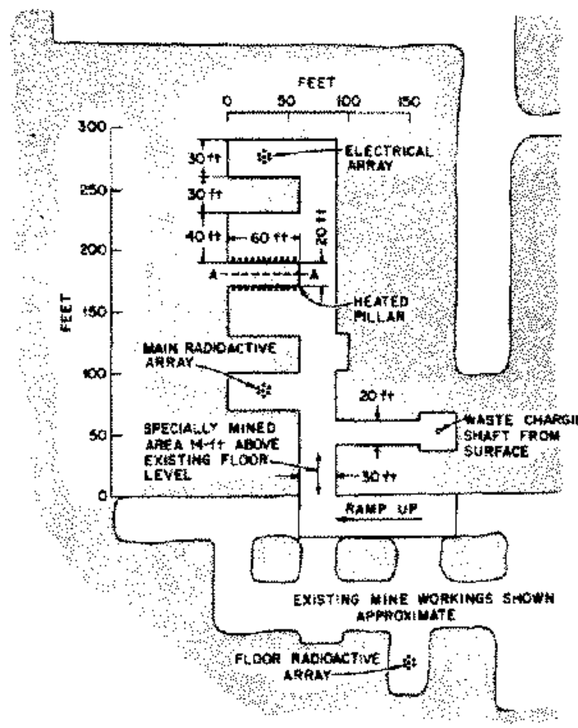

Figure 2. Layout of experimental area in Carey Salt mine, Lyons, Kansas (from Empson, et al., 1966).

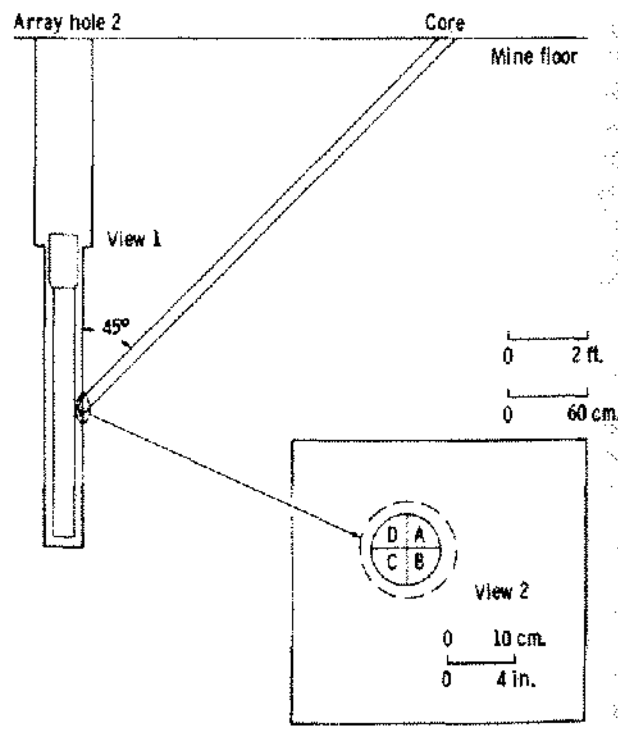

Figure 3. Cross section through part of array hole and core.

bleaching or change in color occurred during the preparation of thin sections or during petrographic studies.

PETROFABRICS

In thin section, rock salt is colorless, isotropic, and shows low relief. Salt from Lyons is composed of anhedral grains of halite from 5 to 30 mm in diameter. The grains may be clear or cloudy, depending on the number of fluid inclusions. The main impurity is anhydrite; in addition small amounts of clay minerals may be present.

Coloration of salt

It is well known that irradiation of salt results in coloration of the crystals. In salt irradiated in situ during the Project Salt Vault experiment, the radiation-induced color varies with distance from the array hole. In the core examined, salt nearest the array hole is blue-black in color. Further from the array hole, the intensity of the blue coloration gradually decreases, the salt being royal blue at 17.5 cm from the array hole. The salt ranges from purple to pale purple between 22.5 and 45 cm from the array hole. Small patches of purple salt occur in the dark blue salt, but blue coloration was not observed in the purple salt.

The blue and purple colors observed in irradiated salt are due to the scattering and absorption of light by particles of colloidal sodium. The formation of these particles was discussed by Przibram (1956, p. 86–90). Light is scattered and absorbed by the colloidal particles, producing shades from pale purple to blue-black. The purple colors are due to the presence of particles between 20–40 mμ in diameter, and were observed between 22.5 and 45 cm from the array hole. The blue colors are due to particles between 40–80 mμ in diameter and were seen between 0 to 22.5 cm from the array hole.

The existence of different sizes of colloidal particles was further confirmed by data from thermoluminescence studies (Holdoway, 1972). Samples of irradiated salt were heated at the rate of 1°C per second and the intensity of luminescence recorded. During the heating of dark blue salt, two main peaks were recorded, one at approximately 400°C, corresponding to bleaching of the dark blue color, and one between 320–360°C, corresponding to bleaching of the purple color. The temperature necessary for bleaching was found to vary with rate of heating, and the distance of the sample from the radiation source. This variation in temperature of bleaching is further evidence that the radiation-induced colors are due to colloidal particles, the size of which depends on the total gamma radiation received by the salt crystals.

During the Project Salt Vault experiment, temperatures did not exceed 200° C at the wall of the array hole, and were considerably less than this further from the array hole. Thus heating does not account for the many bleached areas which are observed in the salt. Other mechanisms which could lead to bleaching of the radiation-induced colors include: 1) recrystallization, 2) oxidation of sodium colloid by oxygen or chlorine radicals, and 3) diffusion of OH^-. The first two mechanisms are of particular importance in this study. Jenks (1972) has shown that the migration of fluid inclusions will bring Cl_2^- or Cl_3^- into contact with colloidal sodium in solution in the brine cavities. A reaction would be expected to occur between these two species, and salt precipitated behind the migrating inclusions should be more or less free of radiation damage.

Przibram (1956, p. 42–43) stated that Haverfield observed color in salt which had been stressed by pressures from 500 to 5000 kg/cm^2. No bleaching was reported to have occurred. The increase in stress in the salt due to heating and irradiation and the preparation of thin sections may have caused the migration of defects to certain areas, such as slip planes, but it is unlikely that any bleaching occurred as a result of stress applied during the experiment or the preparation of samples.

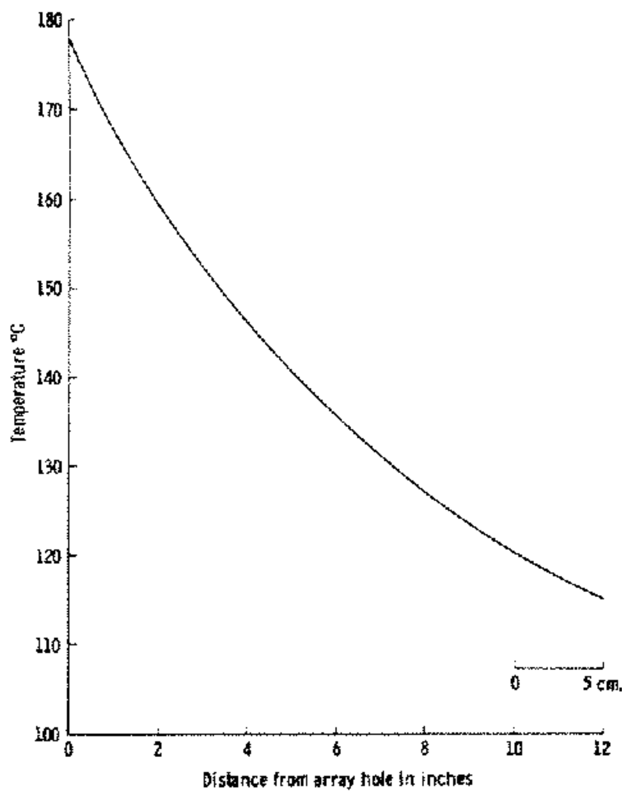

Figure 4. Thermal gradient for salt surrounding array hole 2. All photographs and photomicrographs of irradiated salt are from vertical sections. In photomicrographs, the array hole is situated directly to the right, parallel to the vertical edge of the photographs. In photographs of thin sections, way up is indicated by an arrow. The array hole is situated parallel and to the right of the arrow.

Formation of fluid inclusions

Fluid inclusions occur within negative crystals in the sodium chloride lattice. The cavities are normally cubic, and represent areas where growth did not occur during crystallization. The cavities may not be cubic at the time of formation, but rearrangement generally occurs after crystallization until the cubic form is attained, and they are in equilibrium with the lattice. At the time of formation, the negative crystals are filled with brine. At lower temperatures the liquid contracts, forming a vacuole with a partial vacuum.

Fluid inclusions may be primary or secondary. Those formed during the precipitation of the salt crystals from solution are primary and contain brine from which the salt crystallized. Secondary inclusions are common where crystallization has occurred along cracks and fractures in the salt. The size, abundance, and arrangement of primary fluid inclusions is variable and reflects conditions during crystallization of the salt. Rapid dentritic growth may result in the trapping of numerous small liquid inclusions, whereas slow growth may lead to the trapping of few larger inclusions.

In the core examined from the mine at Lyons, bands of cloudy salt with abundant inclusions and bands of clear salt with few inclusions have been observed. The inclusion-rich bands are infrequent and thin, and show small, numerous inclusions which are often arranged in chevron structures. Such chevron structures represent a primary growth feature of halite crystals and indicate that recrystallization has not taken place. Chevron structures have been observed by Wardlaw and Schwerdtner (1966) in Devonian salt from Saskatchewan. They observed grains with chevron structures elongated perpendicular to bedding which appear to have grown upward from the bottom of the basin. However, chevron structures in the Lyons salt lack orientation, and it is more probable that they represent parts of hopper crystals which grew floating on the surface of the brine in the manner proposed by Dellwig (1955) (Fig. 5). The grains showing chevron structures generally have an inclusion-free rim, suggesting that these grains were formed by fragments of hopper crystals sinking and acting as nuclei for further crystallization. Most growth of salt probably occurred on the bottom of the basin, by crystallization from dense brines, salt crystallizing in this way being relatively inclusion-free. Inclusion-free salt could also be the result of post depositional recrystallization.

Figure 5. Photomicrograph of a thin section showing a pyramidal-shaped hopper crystal from the Salina Salt of Michigan (from Dellwig, 1955). X 19.3

Effects of heating and irradiation on salt containing fluid inclusions

Experimental work. Bradshaw and Sanchez (1968) have shown that the creation of a thermal gradient across salt crystals will cause migration of fluid inclusions. Inclusions with less than ten percent vapor migrate toward the heat source, and those with more than ten percent vapor migrate away from the heat source (Anthony and Cline, 1972). In the sample studied during this investigation, migration appears to have occurred almost exclusively toward the heat source. The solubility of salt increases slightly with increasing temperature. Thus the wall of a fluid inclusion nearest the heat source will be dissolved preferentially by the brine within the inclusion and be precipitated on the wall of the inclusion furthest from the heat source. This mechanism will result in the migration of fluid inclusions up the thermal gradient. Movement would be expected to occur in a straight line towards the heat source, as the effect of the thermal gradient is greatest in this direction. Anthony and Cline (1970) have shown that large inclusions migrate more rapidly than small ones, the rate of migration being controlled by the kinetics of dissolution and deposition at the cavity walls, and that inclusions below a critical size do not migrate.

The migration of fluid inclusions in irradiated salt has been investigated by Jenks (1971). He stated that radiation results in the breakdown of molecules present in brine, with recombination of ions occurring to produce new chemical species such as H_2, O_2, Cl_2, $C10_3^-$, within the cavities. Oxidizing and reducing agents are introduced into the brine as migration takes place. The oxidizing agent corresponds to Cl_2^- and/or Cl_3^-, while the reducing agent is derived from colloidal sodium and can be treated as a hydrated electron, e_{aq}^-. When these species enter solution in a brine cavity, the colloidal sodium may be oxidized and thus dispersed, and colorless salt with few defects may be deposited on the back wall of the cavity. In this way, a bleached trail could form showing the path of migration of a fluid inclusion through irradiated salt.

Bradshaw, et al. (1968) investigated the effect of temperature on salt from Lyons and Hutchinson. They found that salt decrepitates at around 280°C, releasing a cloud of steam. They also found that negative crystal cavities undergo permanent volume expansion on heating, because the expansion of the fluid in the cavities is greater than that of the surrounding salt. The increase in volume is proportional to the temperature, being 7.5 percent at 225°C. Jenks (1971, p. 228) stated that cavity expansion favors the formation of a gas-vapor phase within the cavities.

Evidence for the migration of fluid inclusions. Thin sections from the core reveal many bleached areas in the radiation-colored salt. The bleached areas vary in shape, but in vertical sections many are roughly rectangular in form with their longest dimension perpendicular to the array hole. In horizontal sections only parts of the bleached areas may be seen, thus vertical sections were used predominantly in the petrographic studies. The bleached areas generally have one or more fluid inclusions at the array hole end, and appear to be trails showing the path of migration of fluid inclusions toward the array hole (Fig. 7).

These visible trails could have been formed by the migration of inclusions during 1) heating and irradiation of the salt, 2) the cooling stage of the experiment, or 3) a combination of both 1) and 2). The patterns of bleaching within a grain depend on the size, number, and arrangement of the fluid inclusions. Thus, the primary structure of the salt is largely responsible for the pattern of bleaching observed. Little migration across grain boundaries appears to have occurred. Trails produced by the migration of single, isolated fluid inclusions are observed in all colored parts of the core. The size of a trail depends on the original size of the fluid inclusion, and the distance of the inclusion from the array hole. This dependence makes it difficult to estimate the rate at which migration occurred.

In the cloudy bands of salt as much as half of the salt may be bleached. Bleached areas are crowded with small fluid inclusions, which are frequently arranged in chevron structures (Fig. 6). Small trails may be seen entering or leaving the large bleached patches, with their longest dimension perpendicular to the array hole and a single inclusion at the array hole end. This shows that the direction of migration was directly towards the array hole (Fig. 8).

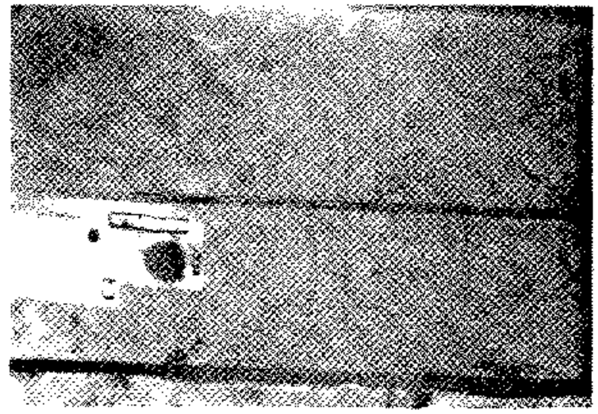

Figure 7. Photomicrograph showing bleached trails produced by the migration of single inclusions and two sets of slip planes (22 cm from the array hole). X 14

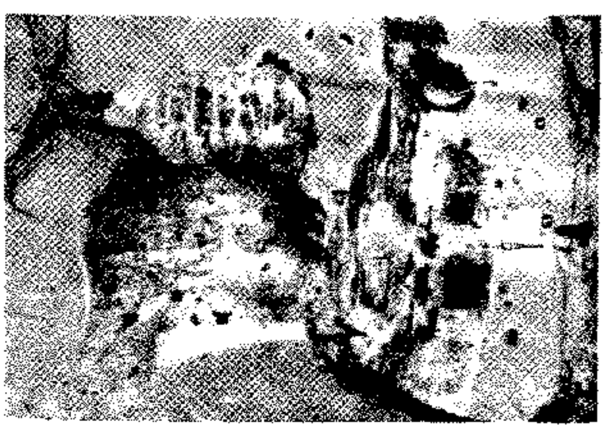

Figure 6. Photomicrograph of a thin section showing relics of chevron structures. Some fluid inclusions appear to have migrated towards the array hole (28 cm from the array hole). X 14

Clusters of small inclusions and streams of larger inclusions occur within the bleached areas. The clusters form remnants of chevron structures. Different migration rates of inclusions, resulting from variations in size and thermal gradient, could readily have produced the observed patterns from chevron structures (Anthony and Cline, 1970). It appears that the larger inclusions in a chevron structure were able to migrate, and moved away from the chevron structure, and very small inclusions were either unable to migrate, or migrated a very small distance, preserving the chevron structure. Immediately adjacent to the array hole, the bleached patches show streams of fluid inclusions. The inclusions are numerous along the length of the trails, but are particularly abundant at the array hole end (Fig. 10). Small inclusions often occur in clusters, particularly at the

Figure 8. Photograph of a thin section showing large irregular bleached areas (25-27.5 cm from the array hole). X 2

Figure 9. Photograph of a thin section showing large bleached areas and secondary anhydrite (top left) (0-4 cm from the array hole). X 2

ends of the trails furthest from the array hole. In some cases, relics of chevron structures may be seen, suggesting that the streams of inclusions inside the bleached areas may be derived from chevron structures. These structures could have been destroyed by the migration of constituent inclusions under a high thermal gradient. This theory is supported by the fact that the larger inclusions occur at the end of the trail nearest the heat source, and clusters of tiny inclusions occur at the end of the trail furthest from the heat source. The occurrence of chevron structures in adjacent grains indicates that the inclusions forming the trails probably originated within this area and were not derived from grains further from the array hole.

Clear bands of salt with few inclusions display different patterns of bleaching. In a clear band of salt between 16 and 21 cm from the array hole, large bleached trails occur which show boundaries parallel and perpendicular to the array hole. These boundaries are straight and sharply delineated (Fig. 11). Few inclusions are observed within these bleached areas, and these trails may have originated from the migration of a single fluid inclusion which could subsequently have split up into smaller inclusions and/or have been dispersed along grain boundaries. These trails occur in grains where the walls of the fluid inclusions are parallel and perpendicular to the array hole. The precise orientation of the bleached trails and the inclusions suggests that these grains may have recrystallized due to heating and irradiation.

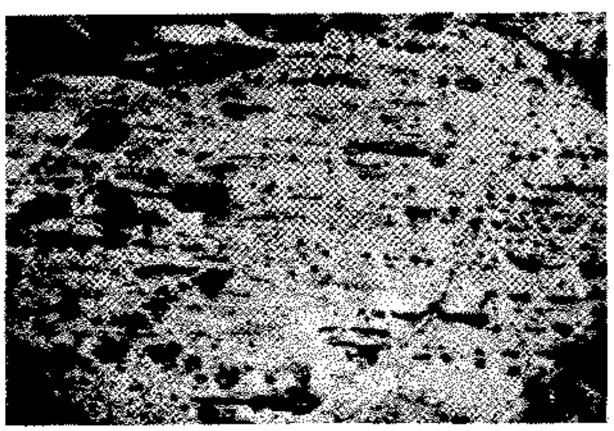

Figure 10. Photomicrograph of large bleached area closest to the array hole (Figure 9) showing partially bleached islands and streams of fluid inclusions. X 14

In a clear band of salt between 12 and 16 cm from the array hole, the bleached areas are very irregular in outline (Fig. 12). The boundary on one side of a bleached patch may be straight and well defined, whereas the boundary parallel to it may be irregular and indistinct. Few inclusions are seen in the bleached salt, or in the surrounding salt. Those that do occur are 0.3 mm or less in diameter and are not usually oriented with respect to the array hole. Cleavage planes in this horizon appear to show some distortion, being offset along their length. The above evidence suggests that some plastic deformation occurred between 12 and 16 cm from the array hole.

An anomalous bleaching pattern with dendritic form was observed throughout the core, but is particularly prevalent in pale purple salt. It is composed of an anastamosing network of thin bleached lines. The network is, in many places, connected by several branches to a grain boundary and also is commonly in close proximity to an anhydrite mass (Fig. 13). In some cases, particularly near the array hole, tiny inclusions are seen along the individual branches of the pattern. The pattern is very irregular, showing no particular orientation, but horizontal sections show that it is three-dimensional. This pattern may have been formed by water molecules migrating away from the grain boundaries. Recrystallized anhydrite implies that water was present as a transporting medium. This would explain the association of anhydrite with the dendritic pattern. Small amounts of water may have been produced from the dehydration of small amounts of clay minerals.

In conclusion, the major patterns of bleaching observed appear to be determined by the stratigraphic position of clear and cloudy bands within the salt. Cloudy bands contain numerous inclusions which are frequently arranged in chevron patterns, whereas clear bands contain relatively few inclusions.

Effect of migration on the shape of fluid inclusions. Fluid inclusions in most grains are not oriented with their cavity walls parallel or perpendicular to the array hole. Migration of these inclusions occurs as in the idealized picture, with salt going into solution at the walls of the cavity nearest the array hole. In unoriented inclusions, this may lead to an apparent rounding of the cavity. Many inclusions appear rounded in thin section. Some of these are larger than surrounding inclusions, and may have formed by the coalescence of several inclusions. Larger

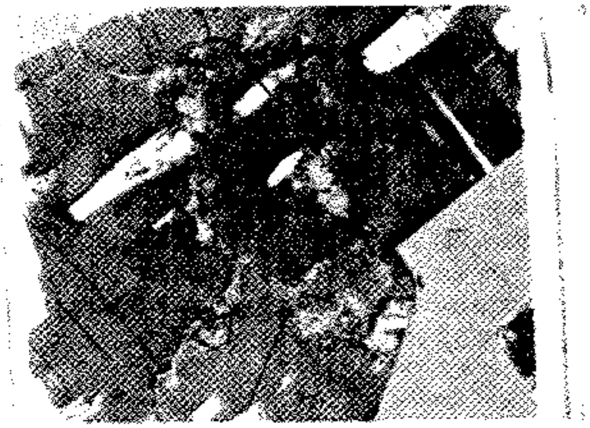

Figure 11. Photograph of a thin section showing bleached trails with straight, regular boundaries. Slip planes are visible in the top left corner. Anhydrite occurs along the grain boundaries (17-21 cm from the array hole). X 2

inclusions migrate faster than small ones and thus may be more susceptible to rounding of the corners. Many unoriented inclusions appear truly cubic, however. These may have migrated more slowly, or returned to the cubic form more rapidly than the rounded inclusions.

Grain boundaries and migrating fluid inclusions. Few trails formed by migrating fluid inclusions are continuous on either side of a grain boundary. In most cases, it appears that the inclusions have not traveled far enough to reach a grain boundary. Of those that do reach a grain boundary, most appear to terminate at this surface; only a few cross into the adjacent grain. Some of the large trails terminate at a grain boundary and, as discussed above, the fluid inclusions may have been dispersed along this surface (Fig. 11).

The common occurrence of primary chevron structures and the close association of streams of inclusions which

Figure 12. Photograph of a thin section showing irregular bleached trails. Secondary anhydrite occurs in radiating clusters (12-15 cm from the array hole). X 2

appear to be derived from these structures suggest that there has been little migration of inclusions across grain boundaries. Cline and Anthony (1971) have examined the thermal gradient necessary for a migrating fluid inclusion to overcome grain boundary tension forces. The thermal gradient from array hole 2 into the surrounding salt is shown in Figure 4. Unfortunately, in areas where the thermal gradient was high enough for migration across grain boundaries to have occurred, the grain boundaries are difficult to distinguish due to numerous cracks formed during cooling after the experiment, and large amounts of anhydrite filling such cracks (Fig. 9).

Interpretation of chronology of bleaching and migration. The migration rates of fluid inclusions have been calculated by Bradshaw (1971) and Jenks (1972). As explained above, difficulties were encountered in measuring the length of bleached trails and therefore in estimating migration rates of inclusions. It seems probable that the bleached trails represent the total distance of migration of inclusions from the beginning of Project Salt Vault until ambient temperatures were attained after the cooling phase of the experiment.

Figure 13. Photomicrograph showing the dendritic bleached pattern associated with a grain boundary and secondary anhydrite (20 cm from the array hole). X 2

It was thought that evidence for the migration of inclusions during heating and irradiation would not be visible in thin sections, as it seemed likely that any bleached salt formed during irradiation would immediately be recolored. However, tiny inclusions forming remnants of chevron structures appear to show little, if any, evidence of migration during the experiment. The close proximity of the chevron relics and the streams of larger inclusions suggests that migration across grain boundaries has occurred infrequently, and therefore the trails seen may represent almost the total distance migrated by inclusions since the beginning of the Project Salt Vault experiment.

The implication is that either little or no migration of the inclusions occurred during heating and irradiation, or that recoloring of bleached salt did not occur.

As stated above, Jenks (1972) has noted that the radiolysis of brine may be expected to lead to the formation of such species as OH^-, H^+, ClO_3^-. These radiolytic products may have been trapped in salt which was recrystallized in the path of migrating inclusions, and may have prevented colloidal sodium from forming again during further irradiation. In this case, recrystallized salt would remain colorless. Gaseous species such as H_2, O_2, and Cl_2 could also have formed. Separation of a gaseous phase would probably not have occurred unless there was an increase in volume of the cavities. Bradshaw (1968) and this study showed that such expansion probably occurred due to heating and irradiation of the salt, and it is therefore likely that some separation of a gaseous phase occurred during irradiation (Fig. 14).

Anthony and Cline (1972) have shown that the presence of less than 10 percent gas in a fluid inclusion will permit migration of the inclusion toward a heat source. The presence of more than 10 percent gas will lead to migration away from the heat source. Bradshaw (1968) estimated that at 225°C cavities had expanded 7.5 percent in volume. It is therefore possible that gases produced during irradiation may have reduced the rate of migration of fluid inclusions toward the array hole during the Project Salt Vault experiment and, in some cases, may have caused some migration away from the array hole.

Stress effects

Expansion produced by irradiation and heating of salt produced an increase in stress around the array hole. It is possible that some of this stress could have been relieved by closure of the hole. Experimental equipment occupied the central part of the array hole, and room was left for this anticipated movement. It was estimated that during the experiment the diameter of the 12 inch hole decreased by approximately half an inch (Bradshaw and McLain, 1971). With some relief of stress at the edge of the wall, it is possible that the maximum stress due to heating and irradiation may have occurred a small distance from the edge of the hole. This may correspond to the area showing slight plastic deformation.

Agullo-Lopez and Levy (1964) have shown that gamma irradiation alters the mechanical properties of salt. The main effect is radiation embrittlement, and an increase in yield stress with increasing gamma-ray dose. During the preparation of thin sections, it was apparent that irradiated salt is more brittle than unirradiated salt.

Slip planes. Agullo-Lopez and Levy (1964) determined that in unirradiated salt crystals, almost all glide occurs on one slip system. In irradiated crystals, slip occurs along two perpendicular systems, greatly reducing pile-up. Slip occurs preferentially on dodecahedral planes, thus there are six possible directions along which it may occur.

Slip planes are observed in some thin sections (Fig. 7). Orientation of slip planes varies from grain to grain and is crystallographically controlled. Only certain grains exhibit slip planes, and it is possible that 1) the intense color of the salt near the array hole may mask slip planes in some cases, or 2) that slip planes only develop in certain areas, specifically in grains with a favorable orientation. The most consistent orientation and the greatest development of visible slip planes occurs between 16 and 25 cm from the array hole. Adjacent grains in this area show slip planes with similar orientations. Further, the grains with slip planes exhibit bleached trails and fluid inclusions oriented more precisely than is usual with respect to the array hole. Shlichta (1968, p. 609) reported that badly deformed crystals recrystallize at room temperature by strain-anneal recrystallization in a few weeks. Recrystallization of grains in response to stress would explain the unusually precise orientation of the features described. Slip planes are not visible between 12 and 16 cm from the array hole, and some plastic deformation is thought to have occurred in this area, because the bleached trails are offset along their length.

During differential thermal analysis (DTA), it was found that a large amount of stored energy was released from samples 15 cm from the array hole (Dreschhoff, pers. comm., 1972). It is known that defects may migrate to regions of dislocations (Agullo-Lopez and Levy, 1964). Therefore, it is possible that the defects resulting from radiation could become concentrated in areas where the combined effects of radiation and stress were greatest.

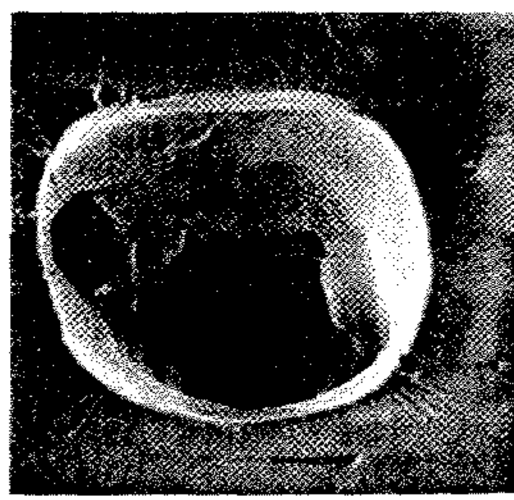

Figure 14. Scanning electron microscope photograph of an etched cleavage surface of irradiated salt showing a negative crystal cavity. Fractures radiate from the surface of the cavity into the surrounding salt. X 300

Impurities

The main impurity is anhydrite. This is most abundant adjacent to the array hole, where it may occupy 25 percent of the area in thin sections studied (Fig. 9). Along the rest of the core, the abundance of anhydrite is variable. It occurs in clumps and along grain boundaries. Secondary crystals are abundant, particularly in the salt close to the array hole. The crystals are long and fibrous, occurring in radiating clusters. They occur around the margins of clumps of primary crystals, along grain boundaries and along fractures. These crystals are frequently seen to penetrate the surrounding halite crystals.

The occurrence of anhydrite along grain boundaries was studied with particular interest, as observations from thin sections suggested that some movement of anhydrite along grain boundaries may have occurred. Problems in the recrystallization of anhydrite over a relatively short period of time involve its low solubility in water. It has been shown, however, that anhydrite is more soluble in brine than in water, and that temperature has only a slight effect on its solubility (Marciniak, *et al.*, 1960). Furthermore, the solubility of anhydrite is sensitive to pressure. Blount and Dickson (1969) have shown that increased pressure causes an increase in the solubility of anhydrite at all temperatures and sodium chloride concentrations. It is possible that anhydrite could have dissolved in brine occurring along grain boundaries, and subsequently have migrated and recrystallized.

After the completion of the Project Salt Vault experiment and the removal of the radiation source from the array hole, a white powdery substance, believed to be anhydrite, was found in many places on the wall of the array hole. An abnormally large amount of anhydrite occurs in salt adjacent to the array hole. The anhydrite here is nearly all in the form of secondary crystals and occurs along fractures (Fig. 9). Farther from the array hole, the volume of anhydrite is much smaller. Secondary anhydrite 2.5 cm from the array hole displayed no thermoluminescence on heating, suggesting that this anhydrite crystallized after the removal of the radiation sources. This evidence supports the theory that migration of anhydrite occurred during the experiment.

CONCLUSIONS

Heated and irradiated salt from Lyons shows shades from dark blue-black near the array hole to purple and pale purple farther from the array hole. The colors observed in irradiated salt are due to a dispersed metallic sodium colloid. The size of the colloidal particles depends, among other factors, upon the total dose of radiation received, and therefore decreases with increasing distance from the array hole. Data from thermoluminescence show that two preferred sizes of colloidal particles exist. The blue-black color is due to the presence of the larger particles, and the purple colors to the smaller particles. In the dark blue salt, particles of both sizes may occur, but in the purple salt, only the small particles are present.

The majority of bleached areas seen in the radiation-colored salt are a result of the migration of fluid inclusions toward the array hole. It was found that bleached salt is recolored by irradiation less readily than colorless salt which has not been previously irradiated. It is possible that radiolytic products may have been trapped in the recrystallized salt and prevented colloidal sodium from forming again. The relationship between the fluid inclusions forming bleached trails and the primary structures remaining in the salt suggests that the trails seen represent the total migration of the inclusions since the beginning of Project Salt Vault. Radiolysis of brine within the inclusions may have lead to the production of gases which could have slowed down or even reversed migration.

The effects of stress due to heating and irradiation were studied. Evidence for dislocation and gliding was seen in slip planes. These were found to be well-developed from 9 to 12 cm and 16 to 25 cm from the array hole. The orientation of these is controlled by the crystallographic orientation of the salt crystals.

The region at 15 cm from the array hole shows some evidence of plastic deformation. Bleached trails and cleavage planes in this area appear to be deformed. Differential thermal analysis shows that a large amount of stored energy also occurs at 15 cm from the array hole. This region may therefore represent the zone where the combined effect of stress and radiation was greatest.

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