LBL-14080

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Materials & Molecular Research Division

Submitted for the Western Regional American Nuclear Society Student Conference, Oregon State University, Corvallis, OR, March 28-30, 1982

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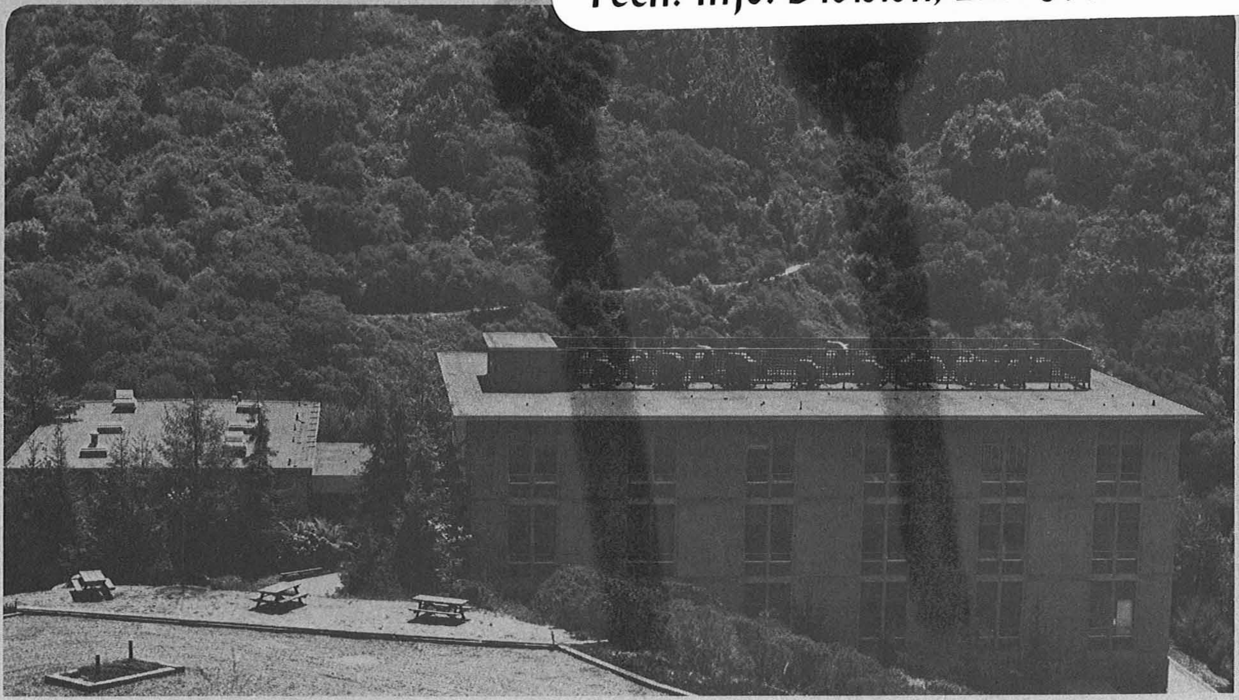
February 1982



Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

THERMAL GRADIENT MIGRATION OF BRINE INCLUSIONS IN SALT

by

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INTRODUCTION

It has been proposed that the high level nuclear waste be buried deep underground in a suitable geologic formation. Natural salt deposits have been under active consideration as one of the geologic formations where a nuclear waste repository may be built in future. The salt deposits, however, are known to contain a small amount ($\sim$ 0.5 vol.%) of water in the form of brine inclusions which are dispersed throughout the medium. The temperature gradients imposed by the heat generating nuclear waste will mobilize these brine inclusions. Therefore, it is important to know the rate and the amount of brine accumulating at the waste packages to properly evaluate the performance of a nuclear waste repository.

MECHANISM OF INCLUSION MIGRATION

Figure 1 shows a photomicrograph of brine inclusions in a salt crystal. There are two types of inclusions normally found in natural salt - all liquid inclusions and gas liquid inclusions. The former ones are completely filled with brine whereas the latter ones have an inert gas (such as air) trapped inside the contained brine. Under an imposed temperature gradient, the all-liquid inclusions move up the temperature gradient towards the hotter regions. The gas liquid inclusions, on the other hand, move down the temperature gradient - from hot regions to cold regions.

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract DE-AC03-76SF00098; and the Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, Ohio.

XBB 823-1982

Fig. 1: A photomicrograph of All Liquid and Gas Liquid Inclusions in Salt Crystal

All liquid inclusions migrate by the diffusional transport of dissolved salt from the hot face of the cavity to the cold face. The solubility of salt increases with temperature, so in a temperature gradient salt dissolves into the inclusion at the hot surface and crystallizes out at the cold surface. The transport through the cavity occurs by molecular and thermal diffusion (Fig. 2). Detailed theoretical model and the migration velocity of inclusions based on this mechanism are given elsewhere[1].

The mechanism of thermal gradient migration of gas liquid inclusions is shown in Fig. 3. Water evaporates from the hot side of the gas bubble in the brine and is transported to the cold side where it condenses. The condensed water is recycled to the hot side by backflow of brine, which provides the mechanism of moving salt from the cold face of the inclusion to the hot face. Olander et. al[2] have given an approximate analytical expression for the migration velocity of gas liquid inclusions.

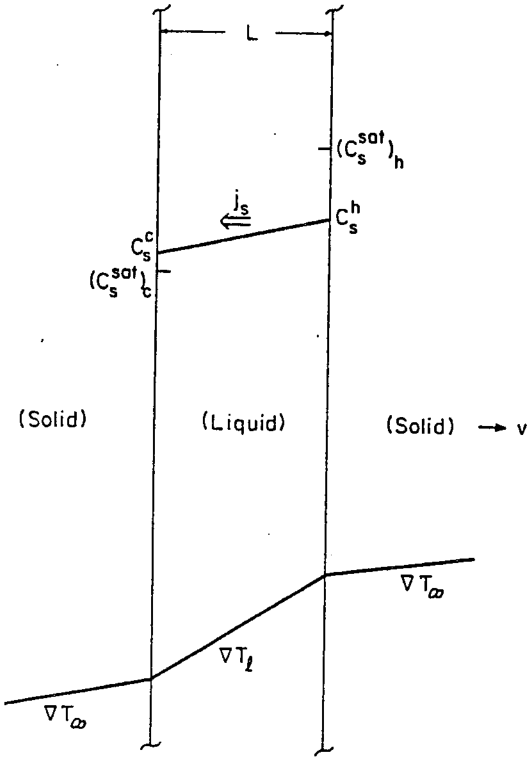

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Fig. 2: Migration Mechanism of All Liquid Inclusions

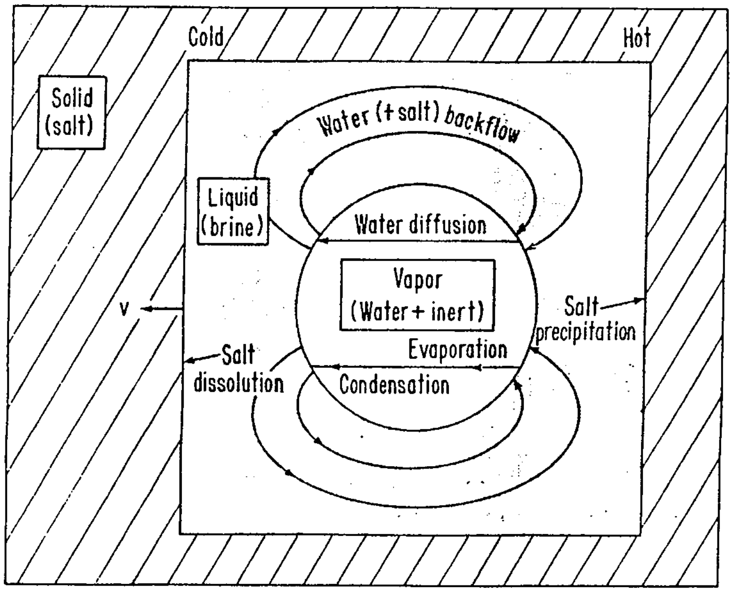

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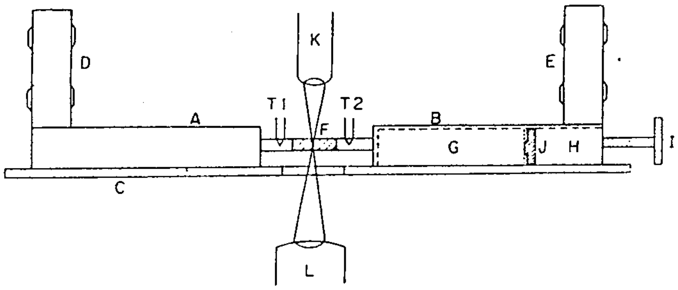
Fig. 3: Migration Mechanism of Gas Liquid Inclusions

EXPERIMENTAL

The experimental setup used to study the behavior of brine inclusions in salt single crystals shown in Fig. 4 is a transmitted-light microscope equipped with a hot stage attachment which maintains the temperature profile in the salt while applying a constant uniaxial stress to the specimen. The hot stage attachment consists of two rectangular copper blocks A and B fixed to insulator plate C. The copper blocks are heated externally by heaters D and E and clamp the salt crystal F between the end faces, which have been polished and gold-plated to promote good thermal contact. The temperatures close to the crystal end faces are measured by thermocouples T1 and T2. Block B is actually a hollow shell with two sliding copper pieces G and H, the movements of which are controlled by knob I. A miniature load cell J is placed between G and H and the axial load on the specimen is measured by the transducer of the load cell.



A, B	Copper electrodes	I	Knob to control load
C	Insulator plate	J	Load-cell
D, E	Clam-shell heaters	K	Microscope objective lens
F	Crystal	L	Microscope condenser
G, H	Inner copper blocks	T1, T2	Thermocouples

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Fig. 4: Microscopic Hot Stage for Migration Studies

Synthetic single crystals of NaCl and KCl with dimensions of 15 mm x 5 mm x 5 mm with (100) faces were obtained from the Harshaw Chemical Co. Inclusions are produced in the synthetic crystal by drilling a 0.3 mm diameter hole 3 mm deep in an end face and filling the hole with deionized water. The crystal is placed between the copper blocks of the hot stage with a sheet of rubber covering the end with the hole to prevent evaporation of the water. By applying a temperature gradient of 10-30°C per cm for several days, the water-filled cavity spawns a cloud of tiny all-liquid inclusions with a size distribution ranging from 10 to 300 μm. These inclusions are initially cubical with (100) planes as faces. When a temperature gradient is applied in a <100> direction, the inclusions move towards the hot end and at the same time change to square platelets which are flattened in the direction of the thermal gradient.

A typical observation consists of measuring the dimensions of the inclusions (thickness L and width W) and the distance travelled during the time between successive observations. With the setup described above, these quantities can be measured with an accuracy of ± 1 μm. To avoid errors due to thermal expansion or creep of the salt, inclusion positions are measured relative to a reference imperfection on the crystal surface. Migration is reversible in the sense that changing the direction of the temperature gradient causes the inclusions to move in the opposite direction.

RESULTS AND DISCUSSIONS

1. Synthetic Single Crystals

According to the BCF theory[3], the rate of removal of solid from the hot face of the inclusion depends upon the presence of dislocations intersecting this surface. Dissolution steps are provided by both screw and edge dislocations. Assuming a dislocation density of 10^5 cm^{-2} and an inclusion 50 μm wide, the average number of dislocations intersecting the dissolving face is 2.5 The number and the location of the dislocations intersecting the hot face of the inclusions are believed to be responsible for the average velocity under specified conditions as well as the dispersion about this average value.

Tests were made to distinguish between control of the migration velocity by liquid diffusion and by interfacial kinetics.

In these tests, several inclusions were selected and the applied temperature gradient ∇T_∞ was varied while keeping the temperature in the vicinity of the inclusions constant. Each time ∇T_∞ was changed, the inclusions changed shape, as expected. For each ∇T_∞ and inclusion shape, the centerline ∇T_ℓ was calculated. For interface kinetic control, the velocity should depend upon the product $L\nabla T_\ell$ raised to some power other than one.[3]

Figure 5 shows the velocities of four inclusions (F, G, H and I) as the applied temperature gradient was varied at constant temperature and low axial load. The same four inclusions were then monitored at axial loads of about 4MPa. As expected from the dislocation arguments presented here, the velocities are higher at the higher load, attaining from 60 to 80 percent of the diffusion-controlled limits. In addition, the velocities are proportional to $(L\nabla T_\ell)$ at high axial load and to $(L\nabla T_\ell)^2$ at low axial load. This load response of the inclusion velocities is in excellent agreement with the model. At low loads the dislocation density in crystal is small and there are few dislocation intersections with the dissolving face of the inclusions. At high loads the dislocation density is increased by deformation and more dislocations intersect the inclusion, making dissolution of the hot face easier. As a result of the reduced interfacial kinetic resistance, the inclusions move at nearly the liquid diffusion-controlled rate.

2. Polycrystalline Natural Salt

The migration studies in high purity single crystals provide an understanding of the basic mechanisms. Natural salt, however, is polycrystalline with high levels of impurities. Thus migration studies were also made on the machined samples of natural salt.

Compared to inclusions in a synthetic single crystal, a higher temperature gradient was needed to cause observable intragranular migration in natural salt samples. This is in agreement with the crystal growth/dissolution theories in the presence of impurities.[3] The high level of impurities in natural salt tend to slow down the dissolution of salt into the inclusion.

The behavior of both the types of inclusions was observed at a large angle grain boundary in natural salt. Under large

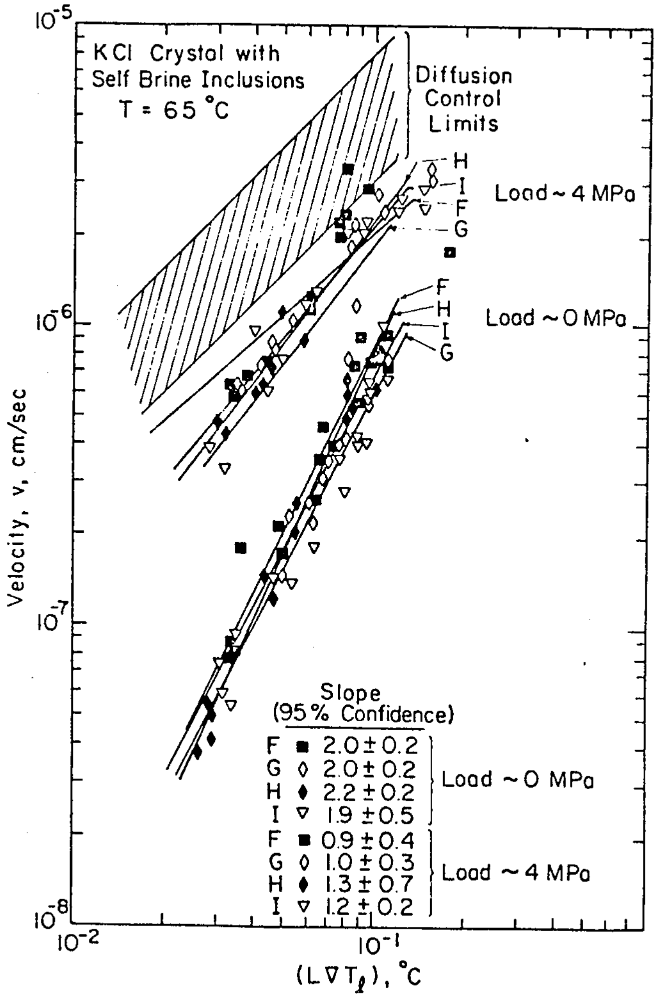

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Fig. 5: Inclusion Velocity Data for All Liquid Inclusions at Low and High Applied Loads. The Velocity is Diffusion Controlled at High Loads but it is Controlled by Interfacial Kinetics at Low Loads

temperature gradients, the inclusions clearly moved past the grain boundary.

The average grain size in salt is about 1 cm. Therefore, a typical natural salt sample for migration study is large (25 x 10 x 10 mm) and semi-transparent. This poses some optical limitations on the microscopic hot stage. A radioactive tracer technique is instead used for migration studies in natural salt samples.

CONCLUSION

An extensive experimental investigation of the migration velocities of brine inclusions in synthetic single crystals of NaCl and in polycrystalline natural salt crystals has been conducted. The results show that in a salt repository the brine inclusions within a grain would move with the diffusion controlled velocities. The brine reaching a grain boundary may be swept across, if the thermal gradient is high enough.

Grain boundaries in polycrystalline rock salt are apparently quite weak and open up due to drilling the hole for a waste canister and to the thermal stresses which accompany the thermal gradient produced by the heat generating waste. The enhanced porosity allows the water reaching the grain boundary to escape by a vapor transport process.[4]

ACKNOWLEDGMENT

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, and the Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus, Ohio.

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