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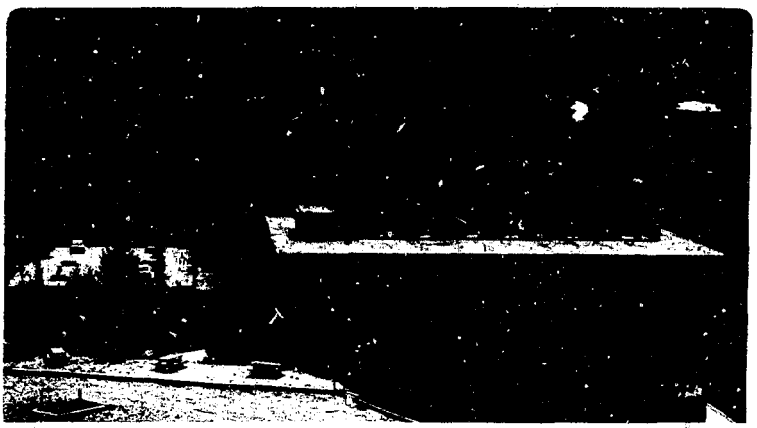

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The liquid content of natural salt is mobilized in the temperature gradient produced by emplacement of nuclear wastes in the deposit. Grain boundaries in polycrystalline natural salt are apparently quite weak and open up due to the thermal stresses which accompany the thermal gradients, allowing water contained in the grain boundaries to be released by a vapor transport process [1]. The intragranular brine contained within the salt crystals migrates in the temperature gradient by the same mechanism by which pores move in nuclear fuels, namely by diffusional transport of the dissolved salt from the hot face of the cavity to the cold face. Upon reaching a grain boundary, the inclusions lose most of their liquid, which escapes by evaporation [2]. Although intragranular water is rapidly lost via high-permeability grain boundaries, intragranular water must first be transported to the grain boundaries to escape. Within a typical grain size of 1 cm, the latter step may control the rate of water loss from rock salt surrounding a waste canister. Consequently understanding of the mechanism of inclusion mobility in salt crystals is important.

Previous experiments [3] have established two salient features of the migration process. First, the rate controlling step is associated with the kinetics of dissolution at the hot face of the inclusion (and to a lesser extent of crystallization at the cold face) rather than with diffusion in the liquid. Second, the experimental data are very scattered, even for experiments conducted under carefully controlled laboratory conditions on high-purity synthetic single crystals. These two aspects of the migration data are in fact manifestations of the same interfacial phenomenon. The rate of solid dissolution at the hot face of the inclusion depends upon the presence of dislocations intersecting the surface, which

provide the imperfections needed to permit dissolution to occur at small undersaturation of the adjacent liquid. If the hot face of the inclusion were flat on an atomic scale, removal of solid atoms from the perfect surface would be so energetically unfavorable that very large undersaturation would be needed to produce the required dissolution rates. A dislocation which intersects the dissolving surface provides sites at which the atoms of the solid are less tightly bound than in a perfect atomic plane and hence their removal to the liquid phase is correspondingly easier. A number of experiments support this interpretation:

1. When the mechanical load on the crystal is increased, additional dislocations are created; these interact with the inclusions, causing them to speed up. The effect is not elastic in nature, because the high migration speeds persist when the crystal is unloaded.
2. Experiments were performed in which selected inclusions were subjected to various temperature gradients but the average temperature was not changed. In a liquid diffusion-controlled process or one with linear interface kinetics [e.g. the theories of Anthony and Cline [4] and Geguzin et. al [5]] the velocity should be proportional to the temperature gradient. However, we found that the velocity varied as the temperature gradient raised to a power > 2. A theoretical dislocation based model which explains this observation is the Burton, Cabrerra and Frank theory of crystal growth [6] which, when taken in reverse (for dissolution), predicts a dependence of the velocity on the square of the temperature gradient.

3. If a single inclusion is followed for a long period of time, variations of its velocity by a factor of 4 are observed (Fig. 1). This is attributed to the loss and gain of dislocations at the hot face. In the low dislocation density synthetic crystals used in the experiments, each inclusion face is on the average intersected by less than 3 dislocations. Consequently, loss or gain of one dislocation (for example) can alter the velocity by a substantial fraction.

The pattern shown on Fig. 1 can be rationalized in the manner shown schematically in Fig. 2. At time zero, the hot face of the inclusion is intersected by a single dislocation (No. 1) close to the center of the face of the inclusion. In general, the dislocation does not intersect the surface at 90° so that as the inclusion moves, the intersection point moves further off-axis. At off-axis positions, the temperature of the surface (and hence the undersaturation of the liquid) is less than it is close to the axis; hence the inclusion slows down. Somewhere between 8 and 20 hours the inclusion picks up another dislocation, designated as No. 2 in Fig. 2. The three dislocation intersections with the dissolving face result in a several-fold increase in the inclusion velocity. As the intersections of the No. 2 dislocation with the hot face move off-axis, the inclusion again begins to slow down.

The dislocation-controlled migration process is qualitatively consistent with all experiments designed to test the hypothesis. It also explains the very large scatter of the measured velocities of small inclusions in crystals of high perfection. Larger inclusions in natural

salt crystals, on the other hand, are not limited by the number of dislocation intersections at the dissolving face and these inclusions appear to move at the rate predicted by liquid diffusion control [7].

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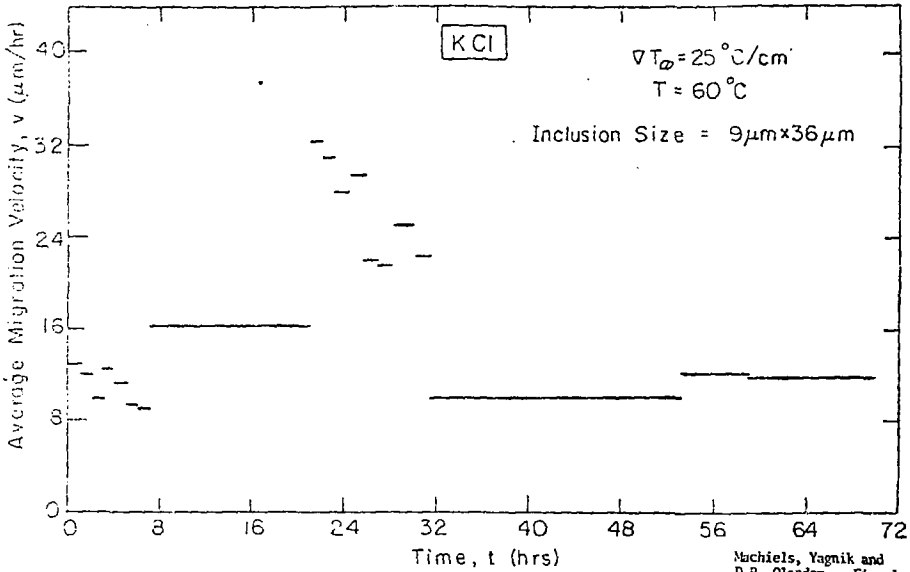
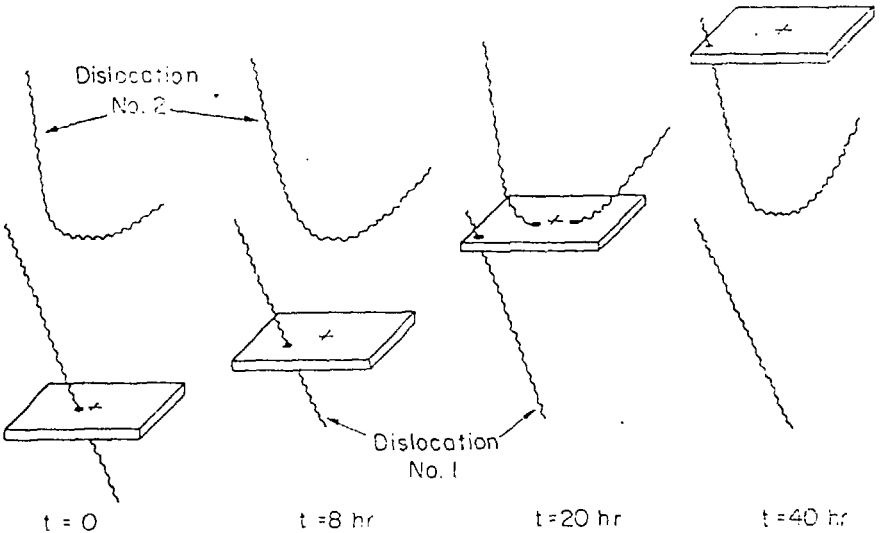


Machiels, Yagnik and D.R. Olander Fig. 1

1. Variation of the Migration Velocity of An Inclusion in KCl



Machiels, Yagnik and
D.R. Olander Fig. 2

2. Schematic Explanation of the Mechanism of the Effect of Dislocation Intersections on the Velocity of Inclusions in Solids