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REPLY TO "REMARKS ON 'MIGRATION OF BRINE INCLUSIONS IN SALT'"

The author is grateful that Chou's[1] remarks about Ref. 2 provide the opportunity to correct typographical errors and to update the evidence for a critical concentration difference, i.e., "critical supersaturation," for brine migration.

Equations (30) and (35) should be corrected as shown in Chou's[1] Eqs. (1) and (2), and the dimension of the second column of Table I should read "$^\circ C^{-1}$."

Chou adopts what is evidently newer solubility and Soret data, though unreferenced, to calculate the threshold temperature gradients at two different temperatures. His results, indicate by lines in his Figs. 1 and 2, differ little from the threshold gradients that this author calculated, shown by the solid circles in these same figures. It is good to learn that the new parameters adopted by Chou have so little effect on the predicted threshold gradients.

The remainder of Chou's comments seems to question that existence of the critical supersaturation ΔC^*. Before responding specifically to his comments, it is more constructive to summarize additional evidence not available when Ref. 2 was first submitted for publication. Olander et al.[3] show experimentally that the aspect ratio of a migrating inclusion in KCl depends on the critical supersaturation ΔC^*, as theoretically predicted by their Eqs. (1) and (A12). From their plots of the variation in inclusion size with applied temperature gradient (Fig. 7 of Ref. 3) at 63°C, Olander et al. deduce finite values of the critical ΔC^* shown in their Table 4. Also, by fitting their migration velocity data to the theoretical nonlinear kinetic model [their Eq. (18)], they deduce values of ΔC^* that generally agree with those deduced from aspect ratio.

Critical supersaturations derived for synthetic single crystals are not necessarily those for natural salt, where impurity cations in the crystals can account for most of the critical supersaturation[4,5] and can be expected to yield higher values of ΔC^* than for pure single crystals. Yagnik[6] observed that inclusions in rock salt were stationary for temperature gradients of 10°C/cm, whereas similar size inclusions in synthetic NaCl single crystals exhibited small but measurable migration at 10°C/cm. Yagnik's data show migration velocity as a function of $L\nabla T$ and indicate the existence of a threshold gradient for migration.

Within the range of these experiments, which were limited to very small inclusions, critical supersaturation is nonzero. Chou's statement that critical supersaturation disappears at high temperature is probably correct at some temperature, because the solid state finally disappears. It is speculative to conclude that critical supersaturation disappears within the temperature range of repository salt, and it is incorrect to infer that critical supersaturation does not exist in the lower temperature regions of a salt repository where it is important.

The theory of inclusion migration summarized in Ref. 2 assumes linear interfacial kinetics [see Eqs. (1) and (2), Ref. 2]. Olander et al.[3] have included a more general description of the interfacial kinetics to encompass the theory of Burton et al.,[7] who conclude that interfacial kinetics are linear when the critical supersaturation is large and are nonlinear when the critical supersaturation is small.

Reference 2 and Chou's comments speak of the migration of brine inclusions *across* grain boundaries, implying that migration proceeds from one halite crystal, across the grain boundary, and into an adjacent crystal. More recent data[6] on brine release from polycrystalline halite suggest that inclusions reaching a grain boundary then migrate along grain boundaries rather than across adjacent crystals. Mobility along these grain boundary pathways can be affected by stress concentrations, including thermal stress, and must be considered in more detailed calculations of the migration of brine inclusions in repositories.

The author agrees with Chou that his calculations show that the 0.1-cm brine inclusions in Waste Isolation Pilot Plant (WIPP) salt will migrate if exposed to the maximum temperature gradients quoted for the WIPP repository, but this does not support Chou's conclusion that most of the liquid in WIPP halite will migrate. The temperature gradients are only temporal, and the maximum gradients occur only in the halite at the surface of the waste package boreholes. Most of the WIPP halite will never experience these maximum gradients, and much of the halite will be below the threshold gradients calculated in Ref. 2. The point of Ref. 2 is that these interfacial kinetics that exhibit critical supersaturation effects do affect brine migration and that they will affect the extent of brine migration in a salt repository. Of course, Chou may elect to ignore interfacial effects for "conservative" estimates, but interfacial effects are clearly important for the most realistic estimates of brine migration in a salt repository.

As further evidence for the existence of a threshold temperature gradient, below which inclusions will not migrate, Ref. 2 showed that without such a threshold the inclusions in the 200-million-year-old halite crystals should have migrated to grain boundaries under the influence of the geothermal gradient. The author agrees with Chou that gravity exerts body forces on inclusions comparable to the forces calculated from geothermal gradients. However, instead of making the argument "invalid," the presence of the inclusions even after such long exposure to gravitational forces is further evidence of a finite migration threshold that can be interpreted as a potential required to nucleate steps on crystalline surfaces bounding a migrating inclusion. Although Chou may infer that under some unique conditions the opposing gravitational and geothermal forces on an inclusion may exactly cancel and result in no migration over geologic time, a small imbalance in these forces would, in the absence of threshold phenomena, result in either one of these forces sweeping the inclusions to the grain boundaries. Examination of the varying geothermal gradients and the existence of inclusions in so many different salt locations confirms the improbability of such a precise balance of forces and further supports the existence of a threshold potential for migration. These considerations of gravity were explored with Jenks shortly after Ref. 2 was published, and the author agrees with the later discussion thereof by Jenks and Claiborne.[8]

Chou's inference that radiation-damage effects are important to the issue of brine migration, particularly under the conditions where threshold gradients are important, is not supported by the reviews of Jenks[9] and Jenks and Claiborne[8] of the knowledge of radiation-damage phenomena in salt.

The author thanks Dr. Chou for his careful and thorough reading of Ref. 2 and for providing the opportunity to clarify and update the more substantive issues on interfacial kinetics and threshold phenomena that affect the migration of brine inclusions.

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July 1, 1983

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