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EFFECTS OF BRINE MIGRATION ON WASTE STORAGE SYSTEMS

Final Report for P.O. 2421409

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ABSTRACT

Processes which can lead to mobilization of brine adjacent to spent fuel or nuclear waste canisters and some of the thermomechanical consequences of that mobilization have been investigated. Migration of fluid inclusions due to thermal gradients is likely to dominate such mobilization. Thermal gradient migration will depend on the size and composition of the inclusions, the temperature and the temperature gradient. Large inclusions should move faster than small ones. High temperatures and high gradients both lead to high migration rates. There is considerable uncertainty in the value of some of the independent variables in the equations governing motion, most notably that of the Soret coefficient. Nevertheless, calculated values for brine accumulation during Project Salt Vault are consistent with observations. Velocities as high as $4 \times 10^{-7} \text{ m s}^{-1}$ (13 m yr^{-1}) are calculated at the salt/canister boundary. Based on calculated velocity fields, as much as 40 liters of pure NaCl brine could accumulate around each canister during a 10 year storage period. Accumulations of bittern brines would probably be less, in the range of 2 to 5 liters. With 0.5 per cent water, NaCl brine accumulation over a ten year storage cycle around a spent fuel canister producing 0.6 kW of heat is expected to be less than 1 liter for centimeter-size inclusions and less than 0.5 liter for millimeter-size inclusions. For bittern brines, about 25 years would be required to accumulate 0.4 liter.

The most serious mechanical consequence of brine migration would be the increased mobility of the waste canister due to pressure solution. In pressure solution enhanced deformation, the existence of a thin film of fluid either between grains or between media (such as between a canister and the salt) provides a pathway by which the salt can be redistributed leading to a marked increase in strain rates in wet rock relative to dry rock. In salt, intergranular water will probably form discontinuous layers rather than films

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so that they would dominate pressure solution. A mathematical model of pressure solution is developed. Using this model, we conclude that pressure solution will not lead to appreciable canister motions except possibly in fine grained rocks (less than 10^{-4} m). In fine grained salts, details of the contact surface between the canister and the salt bed may lead to large pressure solution motions; further study is warranted.

A numerical model of heat and mass transfer in a narrow slot was constructed to simulate the thermal and mechanical effects of a brine layer surrounding a spent fuel canister. The geometry of the assumed brine layer (aspect ratio of 500:1) rendered the problem intractable in terms of computational time. Nonetheless, it is evident that heat transfer in the brine layer is not conduction dominated but has a significant convective component.

1. INTRODUCTION

Considerable interest recently has been directed at the migration of water through salt deposits because of its critical importance to valid demonstration of safe disposal or storage techniques for spent nuclear reactor fuel and high level radioactive wastes. More than twenty years ago the National Research Council determined that massive salt deposits, because of their plasticity, low permeability and dryness, were the best candidate locations for disposal of such materials^[1]. Indeed, there is evidence that the presence of small amounts of water may actually decrease the permeability of salt deposits^[2] (presumably by solution and redeposition which seals cracks which had formerly permitted flow of non-reactive fluids) and, at the same time, increase the plasticity^[3] (presumably by healing pinned dislocations and incipient cracks on the crystal surface). Both phenomena should make natural salt, which often contains up to a few percent water, all the more attractive as a disposal site. Unfortunately, the heat produced by the decaying waste can act to change the situation markedly.

The small amount of water present in most salt deposits occurs as fluid inclusions, or negative crystals (they are usually bounded by {100} crystal faces), ranging in size from submicron to centimeters. Inclusions a few millimeters across are common. Inclusions from both the site of Project Salt Vault in Kansas^[4,5] and the proposed WIPP site^[6] are apparently predominately MgCl_2 - NaCl brines. Should this fluid become mobilized during storage (or after disposal), considerable uncertainty arises as to the degree of isolation afforded by the salt.

1.1 Statement of the Problem

The high temperatures and high thermal gradients in salt adjacent to spent fuel canisters in a storage site can mobilize the brine present in the salt, causing it to accumulate around the canister. Because of the soluble nature of the host rock, this migration will be almost entirely restricted to migration of fluid inclusions through salt grains and along grain boundaries. One goal of the present work is to provide a mathematical model for that migration under conditions applicable to spent fuel storage, and to apply the model to predict brine accumulation rates. This is discussed in Section 2. This report deals with migration through grains; migration along grain boundaries has not been considered.

Once brine has migrated to the emplacement hole, it will alter the environment of the canister. The effect of chemical corrosion is not considered in this work, but we are concerned with the thermomechanical effects of the brine. There will be two principle effects: (1) a continuous layer of brine may be interposed between the canister and the salt which will alter the thermal and mechanical boundary conditions, and (2) the accumulated brine may alter the mechanical response of the salt by "pressure solution" effects which would enhance creep. Work on these thermomechanical parts of the problem is discussed in Sections 4 and 3, respectively.

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2. BRINE MIGRATION

2.1 Approach to Solution

Our approach to the development of a mathematical model for brine migration is based on work already done by others. Concern for the potential mobilization of water adjacent to spent-fuel storage sites, among other reasons, has led to numerous investigations of the motion of fluid inclusions in ionic crystals^[5,7-21]. The most extensive of these have been conducted by T.R. Anthony and H.E. Cline of the General Electric Research and Development Center who studied both thermal gradient^[9-14] and bouyancy^[15,16] induced migration in KCl. Although we are interested in bittern brines ($MgCl_2$ and $CaCl_2$) saturated with NaCl, the processes which will occur should be similar to those in KCl; only the values of numerous parameters should differ.

Anthony and Cline have paid considerable attention to grain boundaries^[11,16,17] and have found that intergranular surface tension makes it extremely difficult for small inclusions to cross a boundary from one grain to another. (For a droplet of volume $7.3 \times 10^{-15} m^3$, they found a gradient of $290 K m^{-1}$ was necessary to achieve penetration of a 15° twist boundary in the (100) plane^[11]; in centrifuge tests, accelerations of about $10^5 m s^{-2}$ were required^[16].) Because of this difficulty in crossing grain boundaries, Anthony and Cline concluded that brine migration due to waste canister induced thermal gradients could not lead to appreciable water transport over dimensions much larger than one grain diameter^[14]; the fluid inclusions would migrate to the grain boundary nearest the heat source where they would remain trapped by surface tension. The possibility that inclusions as small as 10^{-4} across may readily move along grain boundaries, and that even smaller inclusions may move more readily along grain boundaries than through the body of a grain (see page 6), is not included in the estimates given below (sections 2.2, 2.3 and 2.4).

The equation governing migration of a fluid inclusion through salt (or any other soluble solid) subject to a thermal gradient is^[11]

$$v = -D \frac{C_L}{C_S} \left[\left(\frac{1}{C_L} \frac{\partial C_L}{\partial T} + \sigma \right) G_L - \frac{K}{RTL} - \frac{4\gamma \bar{V}_S}{RTL} \right] \quad (1)$$

where v is the velocity of migration, D is the diffusion coefficient, C_S is the concentration of salt in the solid, C_L is the concentration of salt in the liquid, T is the absolute temperature, σ is the Soret (thermal diffusion) coefficient, G_L is the thermal gradient in the liquid, K is a combined kinetic coefficient for dissolution and precipitation, R is the gas constant, L is the length of the inclusion along the thermal gradient, X is the width of the inclusion (perpendicular to L), γ is the intergranular surface tension between adjacent grains of salt, and $\bar{V}_S$ is the molar volume of the solid. Clearly, the size of the inclusions is of paramount importance in determining their velocity in a thermal gradient. In order to illustrate this, let us break Equation (1) into its separate terms as shown in Table 1 and examine each one separately.

The first term, v_T , arises because the equilibrium concentration of salt in the brine will be slightly greater on the hot side of the inclusion than on the cold side. The principal difficulties in evaluating v_T are estimating D and $(\partial C_L / \partial T)_p$ for bittern brines. Little, if any, data is available on the diffusion of NaCl in $MgCl_2$ solutions. Furthermore, we are interested in diffusion at temperatures between 100°C and 200°C; data on diffusion in this temperature range is lacking even in pure, dilute NaCl solutions. (A similar lack of adequate data on bittern brines affects most of the terms discussed below. Therefore, no attempt has been made to estimate quantitatively the behavior of real brines. Rather, we attempt to estimate how pure NaCl brines will behave. Application of this methodology to real brines must await improved data. Of course, the utility of our

results must be viewed in this light.) We estimate D in pure NaCl solutions at temperatures up to 200°C by extrapolating logarithmically in $1/T$ from handbook data^[22] in the range of 0°C to 30°C. For the value of C_L as a function of temperature, we have taken the value of pure NaCl solutions^[23]. Values thus obtained are presented in Table 2. Using these values we have plotted v_T as a function of temperature for several values of G_L between 10 and 1000 K m⁻¹ (0.1 to 10°C/cm) in Figure 1.

The Soret effect causes ions to migrate in a temperature gradient even in the absence of a composition gradient. If the Soret coefficient is known, the second term, v_σ , can be calculated. Here again we suffer from a lack of data for NaCl in bittern brines. Data are available for NaCl^[24] and KCl^[25] solutions of intermediate concentrations (1M to 4M) over a temperature range from 15°C to 45°C. Because no theoretical basis exists, extrapolation of these data to the range of 100°C to 200°C is risky, but we have assumed σ is independent of concentration and extrapolated both sets of data linearly in T and $1/T$ to put bounds on the expected behavior of σ at high temperatures. At 200°C the four values extrapolated differ by as much as a factor of 3. We have designated the $1/T$ extrapolation of NaCl data as the "best estimate" because it is intermediate in magnitude. Values of σ , including "best" and maximum and minimum, so obtained are given in Table 2. Using these data, v can be evaluated and is shown in Figure 2. Note that, for NaCl, v_σ is larger than v_T and of the same sign.

The necessity of nucleating a new (100) plane for dissolution (or precipitation) on the advancing (or retreating) face of the fluid inclusion leads to a kinetic frictional force on the inclusion. No data are available on the magnitude of K in NaCl crystals, so we have assumed that $K_{NaCl} = K_{KCl}$ which is reported to be between 0.1 and 2.0 J-mole⁻¹ [9]. We use the intermediate value of 0.5 J-mole⁻¹ independent of temperature, which gives the values plotted in Figure 3 when inserted into the equation for the third term, v_K , in Table 1. Note that v_K is always of opposite sign from the total velocity, v . On a grain boundary this term may vanish.

The final term, v_Y , arises because a fluid inclusion moving away from a grain boundary will produce a finger of the grain on the retreating side of the inclusion into the grain on the advancing side, thereby increasing the area of contact between the two grains. The magnitude of γ , the inter-granular surface tension, will vary from boundary to boundary depending on the relative orientation of the two grains. Good data are again lacking so we have assumed a value of $0.05 \text{ J m}^{-2} \text{ mole}^{-1}$, independent of temperature, based on values for KCl^[16], which leads to the values of v_Y plotted in Figure 4. This term will be non-zero only when a fluid inclusion is leaving a boundary; within a grain or approaching a boundary this term will vanish.

Looking at the equations in Table 1, we see the driving terms, v_T and v_σ , are unaffected by the inclusion size, but vary linearly as the thermal gradient. On the other hand, the retarding terms, v_K and v_Y , are unaffected by thermal gradient, but vary as the inverse and inverse square of the inclusion size, respectively. Furthermore, all terms increase with temperature, but the driving terms increase more rapidly than the retarding terms. Thus, the balance between $v_T + v_\sigma$ and $v_K + v_Y$ will depend critically on the size of the inclusions, the magnitude of the thermal gradient, and the ambient temperature. Values of the net migration velocity (v) for several values of L and for several values of G_x are shown in Figures 5 and 6, respectively. For large inclusions ($L = 10^{-2}, 10^{-3} \text{ m}$), we see that v increases by about two orders of magnitude between 40°C and 200°C . For smaller inclusions ($L = 10^{-4} \text{ m}$) the increase is even more dramatic since $v_T + v_\sigma > v_K + v_Y$ for gradients less than 300 K m^{-1} at 40°C . Very small inclusions ($L = 10^{-5} \text{ m}$) will be stationary even at 200°C . For millimeter size inclusions, the effect of thermal gradient is also quite pronounced. At high temperatures, where size-dependent terms become relatively unimportant, the variation is nearly linear. But as the temperature drops, motion of the inclusions will halt at various gradients depending on the diameter. A one millimeter inclusion will halt in a thermal gradient of 10, (30 or 100) K m^{-1} at temperatures of about 140, (40 or 20) $^\circ\text{C}$, respectively. Thus, the behavior of fluid inclusions will vary considerably depending on the situation in which they are found.

2.2 Application to Project Salt Vault

In view of the importance of temperature in determining inclusion migration rates, we might expect that calculations based on values at 40°C ^[14] would be very poor reflections of actual migrations in a waste repository at elevated temperatures. We can combine v_T , v_D , v_K and v_Y for the conditions of Project Salt Vault, a waste disposal demonstration about a decade ago near Lyons, Kansas, to provide an estimate of expected brine accumulation. Holdaway^[18] shows a plot of temperature versus radius from the canister axis in the salt surrounding a canister which has been used to estimate graphically both T and ∇T . The thermal gradient in the liquid G_L will be about $1.5 \nabla T$ ^[14]. Using these values, and interpolating in Figures 1 through 4, we calculate the expected radial dependence of the brine migration rate in Project Salt Vault (subject, of course, to our assumption of properties as estimated, that is, a pure NaCl inclusion). This rate is plotted for three inclusion sizes in Figure 7, where we see that the region of high migration velocities is restricted to about 0.5 m of the edge of the emplacement hole. (We have assumed here that the inclusions contain no vapor phase. If vapor phase is present in inclusions to an extent exceeding about 10 percent, Anthony and Cline^[12] have shown that the direction of motion of inclusions may be reversed at temperatures near room temperature. Their behavior at repository temperatures must be considered to be unknown. Holdaway^[18] did not observe any outward migration in Salt Vault.)

The depletion time t_d required for all inclusions of a given size to migrate to the edge of the emplacement hole (at a range R_0) from a range R_d will be

$$t_d = \int_{R_0}^{R_d} \frac{dR}{v(R)} \quad (2)$$

where $v(R)$ has just been given in Figure 7. The volume of this depleted region $V_d(L, t)$ will then be given by

$$V_d = \pi H (R_d^2 - R_0^2) \quad (3)$$

where H is the length of the storage canister. The Salt Vault canisters were about 2 m long, but we have a cylindrical model. To compensate for end effects, we have used $H = 3$ m. The predicted growth of $V_d(L)$ with time for Project Salt Vault conditions described above is shown in Figure 8; these values were obtained by performing the integration of equation (2) graphically in Figure 7.

Canisters were in place for 19 months during Project Salt Vault^[26], and about 120 days were required for cooling the salt around the emplacement holes after the heat sources were removed^[18], for a total duration of about 1.9 yr. However, the heat output of the sources was not constant, so we will use time t_d of 1.6 yr and 1.9 yr to estimate the amount of brine to be expected in the test. The amount of brine we would predict for one emplacement hole in Salt Vault, assuming 0.5 percent of water by weight, is shown in Table 3. The values, ranging from 12 L to 26 L, are considerably greater than those reported for the test^[27]. However, values reported were for steam condensate whereas values calculated are for accumulated brine volumes. When observed condensate values are augmented to account for the probable presence of hydrous crystalline phases^[28], they are consistent with our calculated values.

2.3 Effect of Bittern Brines

As mentioned above, typical fluid inclusions in salt deposits are composed of bittern solutions, rich in $MgCl_2$ and $CaCl_2$ rather than pure NaCl solutions. It would not be reasonable to expect that such brines would have the same properties. We can make some qualitative estimates of how migration velocities will be affected by changes in composition, but the estimates are even less well founded than those derived in the previous section for NaCl solutions.

We see that the NaCl concentration in the liquid (C_L) enters every term in Equation (1). The saturation value of C_L will be lower in bittern solutions which have two Cl for each metal ion. Typically we expect C_L to be about 50 kg m^{-3} . This will cut migration rates by a factor of six. The diffusion coefficient (D) also enters each term. Because the total dissolved solids content of the solution will be somewhat greater for bitterns than for pure NaCl brine, we might expect diffusion be somewhat more difficult; however, this is probably not a large effect. Finally, there is some data on solubility of NaCl in bitterns that can be used to estimate $\partial \ln C_L / \partial T$ which for "WIPP-A" brine^[29] is about 40 percent greater than the pure NaCl brine. Hence, we might expect that the effects of lower diffusion coefficient and higher $\partial \ln C_L / \partial T$ would nearly cancel.

The other terms in equation (1) either are the same for the bittern solutions (e.g. γ which refers to intergranular surface tensions) or provide no basis for estimation (e.g., K and σ). However, unless there are large changes in their magnitude, the concentration effect ($\sim \times 6$) will dominate. In such a case brine migration in real salt deposits would be slowed by a factor of about six relative to pure NaCl brines. Accumulated volumes would be at least as much smaller, perhaps in the range 2 to 5 liters per canister in the case of Project Salt Vault. These volumes are consistent with both the measured condensate^[27] (at the lower end of the range) and the revised estimate of Potter (at the upper end)^[28].

2.4 Application to Spent Fuel Storage

Agreement between the calculation of Sections 2.2 and 2.3 and observed brine accumulations in Project Salt Vault supports the model used. However, there are significant differences between that project and present proposals for interim storage of spent fuel canisters. The principle differences are likely to be (1) that the salt at the proposed WIPP site has only about half as much water as that at Project Salt Vault [32] and (2) that the thermal output of a spent fuel canister will be about 580 watts, less than a third that of the 2000 watt canisters for Project Salt Vault. Correction for the first difference is trivial; it is accomplished by halving the predicted accumulations.

As a part of the calculations described in Section 4, the temperature profile around a 580 watt canister was calculated, as shown in Figure 9. Using the procedure outlined above, brine accumulations can be calculated for this situation. The results are shown in Table 4, from which it can be seen that accumulated brine volumes are from 10 to 100 times smaller than predicted for Project Salt Vault.

3. PRESSURE SOLUTION

3.1 Background

Salt (NaCl) is soluble in water and its solubility is increased by pressure. Thus one might anticipate that, if a solution layer is present between salt and a spent fuel canister, the weight of the canister on the salt might enhance solution and result in macroscopic motions. Deformations of this nature, involving differential solution due to pressure and subsequent migration through a fluid layer, are generally referred to as "pressure solution" in the geologic literature.

The idea of pressure solution was first introduced by Thompson^[33] and was applied to geologic processes almost immediately^[34]. The relation of pressure solution to chemical potential was suggested, in general terms, by the theory of Riecke^[35]. Russell^[36] demonstrated the reality of the process by showing that alum crystals in saturated solution underwent dissolution at stressed locations and redeposition at unstressed locations. Weyl^[37] proposed a phenomenological theory for the mechanism of pressure solution and applied it to deformation of sedimentary rocks. He surmised that material went into solution in an intergranular fluid through which it diffused to low-stress locations. Since that time several workers have shown that pressure solution is an important process in rocks^[38-40].

Quantitative data, especially on rate of pressure solution deformation have not generally been reported. Because of this lack of data, description of pressure solution strain rates has relied primarily on theoretical discourses such as Weyl's^[37]. The principle difficulty with theoretical approaches is the immature state of nonhydrostatic thermodynamics. This area was recently reviewed by Paterson^[41]. Kamb^[42] gives the chemical potential of a solid (nonhydrostatically stressed) dissolving in a fluid in terms of the normal stress at the dissolving surface. This form seems to have been used in all theoretical treatments after that time.

Rutter^[43] and Durney^[44] both describe in detail the mechanism of pressure solution and develop a model for calculating strain rates associated with it. An intergranular film of fluid, perhaps only a few nanometers thick, is presumed to lie between grains. When a load is applied on a grain boundary, a pressure gradient is set up with the highest pressure at the center of the contact area and a flow is established in the fluid. (Because of the thickness of the layer and the high effective viscosity of thin films of water^[45] the flow velocities are very small even with considerable pressure gradients.) The pressure gradient causes a gradient in the concentration of the crystal component in the solution which, in turn, causes migration of that component from regions of high normal stress. Rutter used this model to predict pressure solution strain rates for quartz and calcite. He proposed that pressure solution would be the dominant strain mechanism in saturated monomineralic aggregates of these minerals at low temperatures (900 K for quartz and 700 K for calcite) and low stresses ($<10^{-3}$ G, where G is the shear modulus for both minerals).

Sprunt and Nur^[46] have recently reported that pressure solution rates in many rocks and minerals (novaculite, quartzite, limestone, dolomitic limestone and halite) are proportional to the strain produced by imposed loads. They put an axial compressive load on a rectangular plate of these materials with a circular hole in the center of the plate and exposed the plate and hole to a solution at high pressures and temperatures. All of

their experiments were conducted on materials in linear elastic deformation so proportionality to strain is the same as proportionality to stress. Their emphasis on strain arises from the model of Kamb^[42] in which only the normal stress at the dissolving surface enters the chemical potential. In their experiments the normal stress on the surface of the hole (i.e., solution pressure) was a constant, even though the mean stress in the solid adjacent to the hole varied due to large variations of the tangential stress. For such a situation, the Kamb model predicts a variation of solution rate that goes as the square of the strain.

3.2 Description of Model

A model of pressure solution is derived in a manner similar to that of Rutter^[43] except that we will use a different form for the chemical potential. Consider the situation shown in Figure 10, where a soluble crystal rests on a rigid, insoluble substrate with an intervening thin fluid film. The entire system is immersed in an infinite fluid saturated with respect to the unstressed crystal. We assume that a steady state has been attained whereby the crystal dissolves uniformly along the interface and the fluid film removes the newly dissolved material at a steady rate (which may vary with location) by diffusion. We also assume that the rate is controlled by diffusion rather than by solution kinetics.

For this situation the diffusion equation is

$$\frac{1}{r} \frac{d}{dr} \left(r \frac{dC}{dr} \right) + \frac{A_0}{wD} = 0 \quad (4)$$

where r is the radial coordinate, C is the concentration of salt in solution, A_0 is the (uniform) rate of dissolution, w is the thickness of the fluid layer, and D is the diffusion coefficient of salt in the layer.

The solution to equation (4) in a circle of radius a with a concentration $C(a) = C_0$ is

$$C(r) = C_0 + \frac{A_0}{4wD} (a^2 - r^2) \quad (5)$$

To relate this to the imposed stress σ_a , we note that

$$\sigma_a a^2 = 2 \int_0^a \sigma_z(r) r dr \quad (6)$$

where σ_z is the component of stress normal to the surface*. The stress is related to concentration through the chemical potential, μ ,

$$\mu_{\text{solid}}(\sigma) = \mu_{\text{solution}}(C)$$

or

$$\bar{\sigma} \bar{V} = RT \left[\ln \gamma/\gamma_0 + \ln (C(r)/C_0) \right] \quad (7)$$

*Note that if the stress σ_a is applied over an area πR_0^2 which is covered by n grains, then equation (6) becomes $\sigma_a R_0^2 = 2 n \int_0^a \sigma_z r dr$. But $R_0^2 = n a^2$ so this form is identical to equation (6).

where $\bar{\sigma}$ is the mean stress in the solid, $\bar{V}$ is the molar volume of the solid, R is the gas constant, T is the absolute temperature, γ is the activity coefficient and γ_0 is the activity coefficient in the bulk fluid. To a first approximation, $\bar{\sigma} = \sigma_z + \sigma_r + \sigma_\theta = (2 + 2\nu) \sigma_z$ for a frictionless load on a solid surface where σ_r and σ_θ are the radial and azimuthal stress components in the salt, respectively, and ν is Poisson's ratio (about 0.25 for salt). Using that relation for $\bar{\sigma}$, assuming $\gamma = \gamma_0$ and combining equations (6) and (7) gives

$$\sigma_a = \frac{R T}{\bar{V} a^2 (1 + \nu)} \int_0^a r \ln \left(1 + \frac{A_0 (a^2 - r^2)}{4wD C_0} \right) dr \quad (8)$$

If $1 \gg \frac{A_0 (a^2 - r^2)}{4wD C_0}$, then, noting that $\ln(1+x) \sim x$, we get

$$\begin{aligned} \sigma_a &= \frac{A_0 R T}{4 \bar{V} a^2 (1 + \nu) wD C_0} \int_0^a (a^2 - r^2) r dr \\ &= \frac{A_0 R T a^2}{16 \bar{V} (1 + \nu) wD C_0} \end{aligned} \quad (9)$$

We need not consider $1 \ll \frac{A_0 (a^2 - r^2)}{4wD C_0}$ since that approximation corresponds to very large stresses. Rearranging equation (9) we have

$$A_0 = \frac{16 \bar{V} \sigma_a (1 + \nu) wD C_0}{R T a^2}$$

The solution rate A_0 is related to the relative velocity of approach of the dissolving crystal and the rigid substrate by

$$v = \frac{A_0}{\rho} = \frac{16 \bar{V} \sigma_a (1 + v) w D C_0}{\rho R T a^2} \quad (10)$$

where ρ is the density of the dissolving crystal.

3.3 Pressure Solution for Spent Fuel Storage

Ignoring the electroviscous effect^[43,45] because of the high electrical conductivity of the saturated brine, we can use Elton's calculation of the approach of a round disc to a half space, and estimate that w will be about 10^{-9} m for time of about 10^8 seconds (3 years). In the absence of electroviscosity, the diffusion coefficient will be that of the bulk fluid ($D \sim 3 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$ at 40°C). Other values used in equation (10) will be $\bar{V} \sim 2.7 \times 10^{-5} \text{ m}^3 \text{ mole}^{-1}$, $v \sim 0.25$, $C_0 = 318 \text{ kg m}^{-3}$, $R = 8.3 \text{ J K}^{-1} \text{ mole}^{-1}$, $T = 313 \text{ K}$, $\rho = 2160 \text{ kg m}^{-3}$. Using these values we find

$$v = 0 (10^{-24}) \sigma_a / a^2$$

in SI units.

If we assume $\sigma_a \sim \rho_{\text{canister}} g h_{\text{canister}}$ with $\rho_{\text{canister}} = 7800 \text{ kg m}^{-3}$ and $h_{\text{canister}} = 4.5 \text{ m}$ then $\sigma_a \sim 3.5 \times 10^5 \text{ Pa}$ and

$$v = 0 (10^{-19} / a^2) \text{ m s}^{-1} \quad (11)$$

To find an upper limit for v , let us assume that a is equal to one half of grain size. (This is plausible if we consider that grain boundaries should be more soluble than the bulk of the grain, causing channels to occur along grain boundaries. These channels may only be 10^{-6} to 10^{-7} m in diameter but they would still represent the "bulk fluid" in Figure 10 if w is on the order of 10^{-9} m.)

Figure 11 shows the velocity given by equation 11 plotted against grain diameter. The left hand axis shows the velocity of canister motion while the right hand side shows the integrated displacement over ten years. For grain sizes between 10^{-4} and 10^{-2} m, we would expect displacements between 10^{-2} and 10^{-6} m over 10 years. Displacements in coarse grained salt clearly present no problems even if the acting σ_a is higher by as much as a factor of 10 to 100. However, in finer grained salt deposits, significant displacements could accumulate if σ_a is much over 10^6 Pa. This could be caused by micro-relief on both the salt and metal surfaces. In that case, there would be a high probability of direct contact of canister to salt grain, thereby removing the "intervening" film of liquid. Consequently, the pressure would be transferred direct to solid and any solution effect would be cancelled.

4. THERMOMECHANICAL ANALYSIS OF BRINE ENVELOPE

4.1 NUMERICAL MODEL

In an effort to obtain self-consistent data on the heat transfer and flow characteristics of the canister/brine/salt complex, a free convection analysis of a prescribed brine layer surrounding the canister was carried out. It was assumed that 40 liters of brine was spread uniformly over the lateral surface of a 0.33 m diameter canister approximately 4.5 m high, yielding a brine thickness of about 1 cm. While the physics of brine migration is likely to support brine pockets of more complicated geometry, this model was felt to be as reasonable as any other, considering the dominance of the radial temperature gradient.

Given the geometry of a cylindrical canister (with a heat generation rate assumed to be a constant 580 watts), a cylindrical layer of brine 1 cm thick external to the canister, and a surrounding salt medium, three axisymmetric finite element discretizations were developed and analyzed with the two-dimensional free and forced convection code NACHOS. The first grid consisted of a relatively crude 96-element model, while the latter two used 225 elements. One of these more refined grids concentrated elements at the top and bottom of the brine cavity in order to improve velocity convergence

in the high-gradient regions. The basic element consists of a biquadratic approximation for fluid velocities, a bilinear approximation for the fluid pressure, and a biquadratic approximation for the temperatures. Those portions of the geometry that represent solid material (such as the canister and the salt) are treated such that extremely small velocities are produced.

4.2 MATERIAL PROPERTIES

All material properties used in the thermomechanical model were those of pure NaCl brines. This will lead to some inaccuracies in the results since real brines will usually be bittern brines (large fraction of MgCl_2). However, the qualitative nature of the present calculations does not justify the considerable effort needed to obtain good estimates of properties for bittern brines.

The basic material properties used are shown in Table 5. Those properties not shown in the table are considered to be dependent upon temperature and are supplied through the user subroutines VISCOUS (for viscosity), CONDUCT (for the thermal conductivity), and ISOVOL (for the coefficient of volumetric thermal expansion). The viscosity was determined by the method of Potter^[47]. The thermal conductivity was determined by fitting data of Riedel^[48] on the variation of conductivity of brine with concentration at 20°C and assuming that the ratio of brine conductivity to that of pure water was independent of temperature. The density was taken from a fit to the data of Potter and Brown^[31].

At 20°C, the brine thermal conductivity is $k = 0.57 \text{ W m}^{-1} \text{ K}^{-1}$; the viscosity is $\mu = 0.0015 \text{ Pa s}$; and the expansion coefficient is $\beta = 0.00048 \text{ K}^{-1}$. Based upon these values, the Prandtl number, $\text{Pr} = c_p/k = 11.75$. The Grashof number, $\text{Gr} = \frac{\rho g (T - T_\infty) L^3}{\mu^2}$, is highly dependent upon two unknown factors - the characteristic length, L , and the temperature difference, $T - T_\infty$. If the temperature difference is assumed to be about 20°C and the characteristic length is chosen to be the height of the canister (4.5 m), then $\text{Gr} = 3.6 \times 10^{12}$. This, when combined with the Prandtl number, gives a Rayleigh number ($\text{Ra} = \text{PrGr}$) such that the buoyancy-driven convective flow is highly turbulent, with the laminar region confined to the lower few centimeters of the canister height.

4.3 Free Convection in a Narrow Slot

Estimates of the boundary layer thickness, velocity, and heat transfer coefficient are available from standard texts that describe the boundary layer method. These numbers are instructive in selecting the finite element discretization. For example, based upon the temperature profile

$$T - T_{\infty} = (T_s - T_{\infty}) \left(1 - \frac{y}{\delta}\right)^2$$

and the velocity profile

$$u = u_x \frac{y}{\delta} \left(1 - \frac{y}{\delta}\right)^2$$

where T_s is the canister wall temperature, T_{∞} is the salt surface temperature, u_x is a fictitious reference velocity, δ is the boundary layer thickness, x is the vertical coordinate and y is the coordinate through the boundary layer; the boundary layer thickness and reference velocity are given by [49]

$$\delta(x) = 3.93 \left[\frac{\text{Pr} + \frac{20}{21}}{\text{Pr}^2} \right]^{1/4} \text{Gr}^{-1/4} x$$

and

$$u_x(x) = 5.17 \frac{\mu}{\rho} \left[\text{Pr} + \frac{20}{21} \right]^{-1/2} \text{Gr}^{1/2} x^{-1}$$

Note that the temperature and velocity profiles satisfy the boundary conditions that: (1) $T = T_s$ at $y = 0$; (2) $T = T_\infty$ at $y = \delta$; and (3) $u = 0$ at $y = 0$ and $y = \delta$. Also, the maximum velocity is found at $y = \delta/3$, where $u_{\max} = 4 u_x/27$. Given these conditions, the boundary layer thickness at the top of the canister is found to be of the order of 2 cm, with a velocity of the order of 0.1 m/sec. Considering that the narrow thickness of brine (1 cm) must support a closed convective cell, it is clear that the boundary layer method will be of limited value in estimating the physics of the free convective process for this geometry.

A number of investigators have treated the more difficult problem of free convection in a narrow slot, both experimental^[50] and numerical^[52]. One of the findings is that the heat transfer across the slot, between the hot wall (canister) and the cold wall (salt), is dominated by conduction for situations where the Rayleigh number is "sufficiently small", with the characteristic length in the Grashof number defined to be the slot thickness. For the brine properties given previously, a slot thickness of 1 cm, and an estimated temperature difference of 20°C, the Grashof number is about 3×10^4 . The corresponding Rayleigh number is about 4×10^5 . Elder^[51] defined the "sufficiently small" Rayleigh number to be about 10^3 , implying that the flow and heat transfer for the brine are beyond those for conduction-dominated flows.

Elder^[51] also identifies the situation where $Ra = 4 \times 10^5$ with a single convective cell (although his slot aspect ratio is 20 and that of the brine geometry is 450). This case is typified by three regions (see Figure 12): a wall region at both the hot and cold walls, where thermal gradients are large in the radial direction; a core region, where the radial gradients are small and the axial gradient is uniform; and end regions, where the flow is significantly more complex.

In an effort to capture the character of these flow regions, while keeping the computational costs to a minimum, 24 of the 96 elements for the coarse grid were used for the brine layer. The remainder were used to capture the heat generation and axial thermal gradient in the canister (6 elements), and the far thermal field in the salt (66 elements). For the finer grids, 165 of the 225 elements were used to model the brine, with 15 elements for the canister and 45 for the salt. The large aspect ratio of the slot geometry was also recognized as a debilitating factor, since the flow in the end regions is more difficult to capture. DeVahl-Davis and Mallison^[52] used a finite difference mesh of 101 by 27 for the bottom half of a slot (assuming skew symmetry about the mid-plane) for single cell calculations and an aspect ratio of 10. As the aspect ratio increased from 10 to 20, even a 201 X 27 mesh was inadequate at $Ra > 10^6$. This latter mesh corresponds to a finite element mesh, using the elements described previously, of about 10 elements across the slot and about 200 along the length. For an aspect ratio of 450, even more axial discretization (or at least judicious, non-uniform mesh placement) would be required. Such a mesh would be expensive to analyze, however, the graded 225 element mesh represented an attempt to use a non-uniform grid to aid convergence, while keeping computational costs under control.

4.4 RESULTS

In spite of the above evidence, several attempts to calculate the flow within the brine layer were made. None were completely successful, being divergent because of the inability to capture the reversal of flow in the end regions. The results during the solution iterations are probably meaningful in the mid-slot region, however, and are examined critically. The first iteration consists of a conduction solution of the energy equation and a body-force-driven solution of the radial and axial momentum equations, with the body forces based upon the conduction solution. The second and succeeding solutions of the energy equation will involve convective transport.

The temperature drop for the conduction solution is slightly over 2° , giving a thermal gradient of 215 K m^{-1} . The axial velocity profile accompanying this thermal gradient is shown in Figure 13. The maximum upward velocity is about $.0065 \text{ m s}^{-1}$ and the maximum downward velocity is $-.0040 \text{ m s}^{-1}$. The radial component of velocity is significantly smaller. Similarly the radial pressure gradient is unresolvable, with the axial pressure gradient of about 45 Pa m^{-1} . These results should be used qualitatively only, since a converged solution may have revealed some significant changes.

At the end of a time step of free convection, the thermal gradients tend to flatten near the walls and increase in the core of the flow. At the end of ten hours, for example, the gradient at the canister is 6.5% lower than the maximum, while the gradient at the salt is about 13% lower. The core gradient is about 239 K m^{-1} . The velocity profile tends to shift toward the walls, as well, but the influence of errors propagating from the end regions renders these values qualitative only.

4.5 Discussion

Our choice of 1 cm for the thickness of the brine layer was based on preliminary calculations from Section 2 which indicated 40 liters of saline brine could be expected in Project Salt Vault. However, as pointed out there, much less brine should accumulate about a 580 watt canister. The estimates are less than 0.01 times the value used. These small accumulations would result in layers of brine only about 0.1 mm thick, if distributed over the entire canisters. The difficulties, described just above, that were encountered in numerical modeling of a narrow slot would be considerably aggravated by further increases in aspect ratio. However, it was seen that the mechanical effects of the layer were very small except for the almost complete loss of shear coupling between the salt and the canister; a similar loss would still be anticipated for a millimeter scale layer.

A much more interesting possibility is presented by layers as thin as 0.1 mm, which may not maintain continuity. Because of the finite surface tension of the brine in contact with either the canister or the salt, a thin layer of brine would be likely to break up into discrete beads. Such a situation would lead to only partial loss of shear and thermal coupling across the mixed boundary. This subject is beyond the scope of the present investigation.

Another point that should be mentioned is the likelihood that brine accumulations would not form an even film but rather would collect at the most stable location near the canister. Because of the dependence of the solubility of salt on temperature and pressure, as well as the density difference between brine and salt, it is not immediately apparent where such a location might be.

5. SUMMARY OF CONCLUSIONS

Based on the work reported above we conclude that:

- 1) thermally induced migration of fluid inclusions will be dominant mechanism of brine transport adjacent to a spent fuel canister in salt, in the absence of large fractures;
- 2) fluid inclusion migration in a spent fuel repository will be described by the following equation:

$$v = -D \frac{C_L}{C_S} \left[\left(\frac{1}{C_L} \frac{\partial C_L}{\partial T} + \sigma \right) G_L - \frac{K}{RTL} - \frac{4\gamma \bar{V}_S}{RTL} \right] \quad (1)$$

where v is the velocity of migration, D is the diffusion coefficient, C_S is the concentration of salt in the solid, C_L is the concentration of salt in the liquid, T is the absolute temperature, σ is the Soret (thermal diffusion) coefficient, G_L is the thermal gradient in the liquid, K is a combined kinetic coefficient for dissolution and precipitation, R is the gas constant, L is the length of the inclusion along the thermal gradient, X is the width of the inclusion (perpendicular to L), γ is the intergranular surface tension between adjacent grains of salt, and $\bar{V}_S$ is the molar volume of the solid;

- 3) effects of fluid inclusion size are unimportant for inclusions 1 mm or larger in diameter, except for storage times of more than 10 years;
- 4) the greatest uncertainty in the quantities of equation (1) is the value of the Soret coefficient at high temperatures;
- 5) the time (t_d) required to remove all the water originally between the edge of the emplacement hole (at R_0) and a radius R_d is given by

$$t_d = \int_{R_0}^{R_d} \frac{dr}{v(r)}$$

where $v(r)$ is given by equation (1);

- 6) based on equations (2) and (1), it is expected that between 13 and 26 liters of pure NaCl brine would have accumulated about each canister in Project Salt Vault. A rough estimate for bittern brines is given as about 2 to 5 liters. Although these values are greater than the measured steam condensate, they are consistent when the condensate values are adjusted for vapor pressure effects in the brine;
- 7) accumulations of realistic (bittern) brines about a 580 watt spent fuel canister will be very small with only 0.2 to 0.4 liters accumulating in 25 years;
- 8) pressure solution is unlikely to cause significant canister migrations except, perhaps, in extremely fine grained salt;
- 9) heat transfer through a brine layer is not conduction dominated but has a significant convective component.

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Table 1.

$v = -(v_T + \sigma + v_K + v_Y)$	
SYMBOL = VALUE	EXPLANATION
$v_T = D \frac{C_I}{C_S} \frac{\partial \ln C_I}{\partial T} G_I$	SALT DIFFUSES FROM THE HOT SIDE TO THE COLD SIDE BECAUSE THE CONCENTRATION OF SALT IN SOLUTION IS GREATER ON THE HOT SIDE.
$v_\sigma = D \frac{C_I}{C_S} \sigma G_I$	SALT DIFFUSES EVEN IF THERE IS NO CONCENTRATION GRADIENT BECAUSE THE CHEMICAL POTENTIAL AT CONSTANT COMPOSITION VARIES WITH TEMPERATURE. (IN GENERAL σ MAY BE EITHER + OR -).
$v_K = -D \frac{C_I}{C_S} \frac{K}{nTL}$	THE REQUIREMENT TO NUCLEATE BOTH SOLUTION AND PRECIPITATION WILL IMPEDE MIGRATION.
$v_Y = -D \frac{C_I}{C_S} \frac{4\gamma\bar{V}_S}{RTXL}$	THE INCREASE IN INTERGRANULAR SURFACE ENERGY WHEN A GRAIN BOUNDARY IS CROSSED ALSO IMPEDES MIGRATION.

Table 2. Values of thermodynamic properties of NaCl brines.

Symbol	Quantity(Units)	Value			Source
R	gas constant (J K^{-1})		8.3		-
K	kinetic coefficient (J mole^{-1})		0.5		[9]
γ	intergranular surface tension (J m^{-2} mole)		5×10^{-2}		[16]
T	absolute temperature (K)	313	393	453	-
D	diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)	2.7×10^{-9}	1.3×10^{-8}	2.8×10^{-8}	see text
C_L	NaCl concentration in liquid (kg m^{-3})	318	331	355	[23],[31]
C_S	NaCl concentration in solid (kg m^{-3})	2160	2137	2113	[30]
$(\partial C_L / \partial T)_P$	($\text{kg m}^{-3} \text{K}^{-1}$)	0.187	0.378	0.520	[23],[31]
σ_{max}	Soret coefficient (K^{-1})	2.4×10^{-3}	10^{-2}	1.6×10^{-2}	see text
σ_{best}		2.4×10^{-3}	9×10^{-3}	1.25×10^{-2}	
σ_{min}		1.9×10^{-3}	4.5×10^{-3}	5.9×10^{-3}	
$\bar{V}_S$	molar volume of solid ($\text{m}^3 \text{mole}^{-1}$)	2.71×10^{-5}	2.74×10^{-5}	2.77×10^{-5}	[30]

Table 3. Expected accumulated brine volumes about one Salt Vault canister (in liters) for 0.5% water in salt.

Time(yr)	Inclusion size (m)		10^{-5}	10^{-4}	10^{-3}	10^{-2}
	L=					
0.3			0	7	9	9
1.6			0	12	22	24
1.9			0	13	24	26
3.0			0	14	29	32
10.0			0	14	36	43
30.0			0	14	36	49

Table 4. Expected brine accumulations about a 580 watt spent fuel canister 4.5 m long by 0.3 m diameter.

Time (yr)	L =	Accumulated Brine Volume (liters)			
		NaCl Brine		Bittern Brine	
		1 cm	1 mm	1 cm	1 mm
0		0	0	0	0
5		0.4	0.2	0.07	0.03
10		0.9	0.4	0.15	0.07
15		1.3	0.6	0.22	0.09
20		1.8	0.7	0.30	0.11
25		2.2	0.8	0.37	0.14

TABLE 5
BASIC MATERIAL PROPERTIES

<u>Property</u>	<u>Canister</u>	<u>Salt</u>	<u>Brine</u>
Density at 20°C, $\text{kg}\cdot\text{m}^{-3}$	7800	2159	1041
Viscosity, Pa·s	"SOLID"	"SOLID"	*
Specific Heat, $\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$	400	943	4186
Conductivity, $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	20	*	*
Volumetric Expansion, K^{-1}	"SOLID"	"SOLID"	*

* Property is dependant upon temperature and is provided through a user-supplied subroutine.

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Figure Captions

1. Variation of fluid inclusion velocity component v_T with temperature in salt for several values of thermal gradient in liquid, G_L . This component is independent of the size of the inclusion, L .
2. Variation of the Soret component (v_σ) of the fluid inclusion velocity with temperature in salt for several values of G_L . The bands represent the spread between the maximum and minimum estimates of the Soret coefficient (σ); the line near the top of each band shows the value of v_σ corresponding to the "best estimate" of σ (see text). This component is independent of L .
3. Temperature dependence of the kinetic component (v_K) of the fluid inclusion velocity in salt for several values of L . This component is independent of G_L . Note that the values are negative.
4. Temperature dependence of the surface tension component (v_γ) of the fluid inclusion velocity in salt for several values of L assuming the inclusion is equidimensional (i.e., $\lambda=L$). This component is independent of G_L . Note that the values are negative.
5. Temperature dependence of the net fluid inclusion velocity at a fixed thermal gradient (G_L) of 300 Km^{-1} and various values of L . Inclusions with $L=10^{-4} \text{ m}$ do not migrate in such a gradient at temperatures below about 45°C .
6. Temperature dependence of the net fluid inclusion velocity of an inclusion with $L=10^{-3} \text{ m}$ for several values of G_L . No migration occurs at temperatures below about $25(40,140)^\circ\text{C}$ for gradients below $100(30,10) \text{ Km}^{-1}$.
7. Radial variation of fluid inclusion velocity for three sizes of inclusions about a radioactive waste canister (based on idealized temperature profiles for Project Salt Vault). Vertical line at 0.25 m radius represents the edge of the canister.

8. Accumulation of brine about a single canister similar to those in Project Salt Vault assuming constant heat flux. Inset shows area about the origin (first three years) in greater detail.
9. Estimated steady-state temperature profile around a 580 watt spent fuel canister in salt without a brine layer.
10. Schematic illustration of pressure solution phenomena.
11. Velocity (left axis) of pressure solution migration of a metal canister through salt at 40°C with a contact pressure of 3.5×10^5 Pa as a function of grain diameter. Displacement accumulated over 10 years is shown on right axis.
12. Temperature distribution for $Ra = 4 \times 10^5$ and aspect ratio = 20. Numerical values are $\theta = (T - T_\infty)/(T_s - T_\infty)$.
13. Calculated vertical velocity profile at the midline of a narrow brine-filled slot.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13