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**THE EFFECT OF WATER IN SALT REPOSITORIES:
FINAL REPORT**

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ABSTRACT

Additional results confirm that during most of the consolidation of polycrystalline salt in brine, the previously proposed rate expression applies. The final consolidation, however, proceeds at a lower rate than predicted. The presence of clay hastens the consolidation process but does not greatly affect the previously observed relationship between permeability and void fraction. Studies of the migration of brine within polycrystalline salt specimens under stress indicate that the principal effect is the exclusion of brine as a result of consolidation, a process that evidently can proceed to completion. No clear effect of a temperature gradient could be identified. A previously reported linear increase with time of the reciprocal permeability of salt-crystal interfaces to brine was confirmed, though the rate of increase appears more nearly proportional to the product of $\sigma\Delta P$ rather than $\sigma\Delta P^2$ (σ is the uniaxial stress normal to the interface and ΔP is the hydraulic pressure drop). The new results suggest that a limiting permeability may be reached. A model for the permeability of salt-crystal interfaces to brine is developed that is reasonably consistent with the present results and may be used to predict the permeability of bedded salt. More measurements are needed, however, to choose between two limiting forms of the model.

1. INTRODUCTION

The purpose of this program has been to carry out experiments and develop models that would permit a quantitative treatment of the behavior of water in salt repositories. After some exploratory studies of the behavior of mixtures of salt and brine (Shor et al. 1979), we undertook a systematic study of the consolidation and permeability of polycrystalline salt in brine as a function of time, the applied stress, the initial crystal size, and the temperature (Shor et al. 1981). The results were found to be consistent with an expression proposed by Coble (1961) to account for sintering behavior:

$$\phi(t) = \phi(0) - (p_1/p_2)\ln[1 + p_2 t/Z(0)^3], \quad (1)$$

where

$\phi(t)$ = the void fraction at time t ,

$Z(0)$ = the initial particle size,

p_1 & p_2 = are constants for a given temperature and applied stress.

Then a study was made of the permeability to brine of single interfaces between salt crystals as a function of time, applied stress, fluid pressure drop, and temperature (Gilpatrick et al. 1982). For constant conditions, the total flow of brine, G , showed a linear dependence on the logarithm of time:

$$G(t) = G(0) + (\Delta P/p_4)\ln[1 + (p_4/p_3)t], \quad (2)$$

where ΔP is the hydraulic pressure drop and p_3 and p_4 are constants for a given run. This corresponded to a linear increase with time of the reciprocal permeability of the interface:

$$1/Pr = p_3 + p_4 t \quad (3)$$

Here Pr denotes the permeability for the radial geometry used:

$$Pr = \frac{dG/dt}{\Delta P} \quad (4)$$

Other phases of the program have included studies of the exclusion of brine from polycrystalline salt specimens of various kinds as a function of applied stress and the temperature and studies of the effect of clay (montmorillonite) on the consolidation and permeability of salt crystals.

In this final report, we shall summarize previously reported results and present additional results in each phase of the program. We shall also present a model for the permeability of polycrystalline salt to brine and indicate how it may be used to predict the consequences of the presence of water in salt repositories.

2. EXPERIMENTAL

Details of the experimental procedures and apparatus used in most of the salt consolidation and crystal interface studies have been given previously (Shor et al. 1981; Gilpatrick et al. 1982). Additional apparatus and procedures employed in subsequent work include the following:

2.1 SALT CONSOLIDATION, PERMEABILITY, AND BRINE EXCLUSION

In addition to the columns with diameters of 1.27 and 2.54 cm employed in most of the consolidation measurements, cells were fabricated of monel that could contain cylindrical specimens of salt and brine with diameters of 7.62 and 10.16 cm and could be fitted with a hydraulic ram capable of applying stresses of several hundred bars through a monel piston sealed by rubber O-rings. The salt specimens were either formed in place from a slurry of sized salt crystals in brine or were machined from salt blocks supplied by either the Morton Salt Company or the Hardy Salt Company. In one case, a 7.62-cm specimen was machined from a larger core of natural halite.* Initially the cells were mounted with the cylindrical axis vertical [Fig. 1(b)], but later they were mounted horizontally [Fig. 1(a)].

The movement of the piston was measured as a function of time either with a cathetometer or suitable displacement gauges. Fritted nickel or screens of stainless steel were placed at opposite ends of the specimen. In the vertically mounted apparatus, an annular space with an exit line was located opposite the disk at each end of the specimen [Fig. 1(b)]. When a large volume of brine was being expelled in the earlier stages of consolidation, it was usually collected at the lower end of the specimen (the upper exit line was valved off). In the later stages, the much smaller volume of expelled brine was exposed in the frits or screens to a stream of flowing gas that was then passed through weighable magnesium perchlorate traps. This proved to be an unsatisfactory procedure

*Specimen #87-17 taken at a depth of 898.2 m from drilling AEC-8 near Carlsbad, NM.

Fig. 1. Assemblies for the study of brine migration in polycrystalline salt under stress: (a) horizontally mounted specimens with brine collected directly; (b) vertically mounted specimens with brine collected directly or water collected by flowing gas.

because of plugging of the frits or screens with salt. In later studies, the cell was mounted horizontally. The volume of the brine-collection system at each end of the specimen was reduced to a minimum, and the excluded brine was collected directly throughout the run.

A machined specimen was usually predried to constant weight in a vacuum oven at 150°C and placed in a vessel that was then evacuated. Subsequently, brine was admitted until the specimen was completely immersed, and finally the vessel was opened to the air. The dimensions of the specimen, and the weights before and after the take-up of brine, permitted the calculation of the initial void volume as well as the portion of that volume filled by brine. Following the termination of a

run, the specimen could often be removed intact, dimensioned, weighed, and dried to constant weight in the vacuum oven. This permitted a second calculation of the final void volume and brine content. These calculations were made assuming the specimen was pure NaCl, generally a quite valid assumption. The errors introduced by wiping the specimen dry of brine before weighing were probably the largest. They limited the accuracy of the calculation for the amount of contained brine to about 0.2% of the specimen volume. When salt-clay mixtures were used, it was assumed in calculating the final void fraction that no clay had been lost and that its density was 2.2 g/cm^3 .*

In some runs, an effort was made to determine the effect of a temperature gradient on the rate of brine loss from each end of a specimen. The temperature at each end was maintained by a suitably mounted heater and a control thermocouple inserted through the gas inlet line. The vessel walls were wrapped in thermal insulation to establish an axial temperature gradient as uniform as possible.

As previously (Shor et al. 1981), when pure salt crystals of uniform size were to be consolidated, they were suspended in brine and allowed to settle within the column. This could not be done with mixtures of salt and clay, because the clay settled less rapidly, causing segregation. Instead, the dried salt and air-dried clay were weighed and mixed by gentle agitation. The mixture was placed in the column and confined by the piston before saturated brine was introduced from below. Tests showed this procedure avoids segregation and could be carried out without trapping appreciable amounts of air.

In the earlier stages of consolidation, the permeability of the specimen was estimated as described previously (Shor et al. 1981) by measuring the hydraulic pressure drop and the flow rate of brine. In the later stages, the exit flow rate from the specimen approached the rate at which the brine was being expelled by consolidation. At this point, the input of brine was discontinued, and only the brine expelled from the specimen was measured.

2.2 CRYSTAL INTERFACE PERMEABILITY

The apparatus described previously (Gilpatrick et al. 1982) was modified to permit continuous automatic recording of brine flow and hydraulic pressure drop vs time. This involved the use of a Mettler automatic balance sensitive to 1 mg, an HP-3497A data acquisition unit, and an HP-9735A computer. The cell was modified to a final form (Fig. 2) in which the brine collection ring was reduced in volume and the downstream hydraulic pressure tap was moved from a point in the exit line to the collection ring. The latter change was made to ensure that the measured pressure difference corresponded to the hydraulic pressure drop across the crystal interface and did not include any additional pressure drops in the exit line. The hydraulic pressure drop was measured with a Validyne sensor coupled to the cell through connecting lines filled with dodecane (Fig. 3). It was found necessary to reduce the fluid volume of this pressure-measuring system and to immerse it in the thermostat to avoid the effects of room temperature variation on the small flow volumes of brine (as low as $0.0001 \text{ cm}^3/\text{min}$) being measured. All of the measurements made with this equipment were at $20 \pm 0.02^\circ\text{C}$.

*The density of montmorillonite in the collapsed (dehydrated) form was calculated to be 2.764, corresponding to a d-spacing of $9.6 \text{ \AA}$. In the presence of saturated brine, a water content 0.5 g/g clay and a d-spacing of $15.4 \text{ \AA}$ was assumed, giving a calculated density of 2.246.

Fig. 2. Improved cell for measurement of the permeability of an interface between salt crystals under axial stress.

Fig. 3. Arrangement for brine delivery to and collection from the crystal interface.

3. RESULTS

3.1 KINETICS OF SALT CONSOLIDATION IN BRINE

The results reported previously (Shor et al. 1981) show that the consolidation of salt crystals in brine proceeds to a fairly low void fraction in accord with Eq. 1, an expression that also describes the behavior often observed during the sintering of metal and ceramic powders. The parameters p_1 and p_2 are given by the expressions in Table 1. They include a substantial wall effect, indicated by an observed increase in the void fraction of consolidating columns with distance from the piston and by a dependence of consolidation rate on the column height and radius. The distance through which the stress fell by a factor of e by transfer to the wall was estimated to change inversely as the fourth power of the column diameter. Consolidation proceeded somewhat more rapidly with increasing temperature (to 85°C). This dependence could be represented adequately by assuming p_1 and p_2 to have the same exponential dependence on the absolute temperature (Table 1).

Additional results for salt consolidation are summarized in Table 2 and plotted in Figs. 4 to 6, where they are compared with the behavior predicted from the expressions in Table 1. The parameters p_1 and p_2 were thus set by the experimental conditions; the initial void fraction and the initial average particle size were treated as adjustable parameters. The more recent results include both larger columns of salt, lower final void fractions, and the use of clay as an additive.

3.1.1 Salt Consolidation

Two runs (201 and 202) involved the consolidation of 250- to 420- μm salt crystals in a column 9.54 cm in diameter. In Run 201, the consolidation could be made to agree fairly well with the solid curve (Fig. 4) given by the expressions in Table 1 by assuming an initial average particle size

Table 1. An expression for the consolidation of salt in brine^a

$$\phi(t) = \phi(0) - p_1/p_2 \ln[1 + p_2 t/Z(0)^3]$$

$$p_1(T) = [8.63 \times 10^{-9} + 3.93 \times 10^{-11} \times \bar{\sigma}] \exp[-5130(1/T - 1/293)] ,$$

$$p_2(T) = 3.18 \times 10^{-7} \exp[-5130(1/T - 1/293)] , \text{ and}$$

$$\bar{\sigma} = \sigma(0)(0.77 \times d^6/W) \left[1 - \exp\left(-\frac{W}{0.77d^6}\right) \right] ,$$

where

- ϕ = the void fraction,
- t = the time (min),
- $Z(0)$ = the initial average particle size (cm),
- T = the absolute temperature (K),
- $\bar{\sigma}$ = an averaged effective stress (bar) corrected for the wall effect,
- $\sigma(0)$ = the applied uniaxial stress (bar),
- d = the column diameter (cm),
- W = the weight of salt (g).

^aShor et al. (1981).

Table 2. Additional salt consolidation tests

Run	Applied stress (bar)	Temperature (°C)		Elapsed time ^a (min)	Void fraction	Brine expelled (g)	
		Ram end	Anvil end			Ram end	Anvil end
Run 201—NaCl crystals: ^b 1540 g in 9.537-cm-diam column							
				0	0.251		
	155	20	20	9685 (1)	0.071	(0)	(0)
	155	50	50	12565 (2)	0.065		
	155	80	80	14505	0.041		
	310	84	85	15705	0.015		
	155	76	95	28507	0.013	0.548	18.325
Run 202—NaCl Crystal: ^b 1556 g in 9.537-cm-diam column							
				85	0.159		
	155	20	20	3940 (1)	0.104		
	155	50	50	13690	(0.0367)	(0)	(0)
	155	50	50	17810 (2)	0.0315	0.256	4.080
	155	57	48	18140	0.0278	22.618	6.624
	155	85	67	35225 (3)	0.0024	4.961	18.295
		85	67	48903		15.190	20.625
Run 203—Hardy salt: ^c 1440.3 g, 9.51-cm-diam cylinder impregnated with 53.85 g brine							
				0	0.0727	0.0	0.0
	155	54	68	2650	0.039		
	155	85	64	23482	0.013	14.24	28.452
	155	85	64	39022	0.013	<i>d</i>	31.149
Run 204—Morton salt block: 1610.8 g, 9.50-cm-diam cylinder							
				0	0.115		
	155	20	20	3351 (1)	0.074	50.360	
	155	65	87	4841	0.060	50.481	
	155	64	79	7671	0.050	50.483	
	155	70	76	8646	0.048	50.504	
	155	73	73	11006	0.044		
Run 205—Morton salt block: ^c 1121.6 g, 7.62-cm-diam cylinder							
				0	0.091	(0)	(0)
	155	20	20	23060	0.018	42.676	7.533
Run 206—Halite: ^c 1450.8 g, 7.62-cm-diam cylinder							
				0	0.0045	(0)	(0)
	155	20	20	34320	(0.0)	0.802	0.667

^aNumbers in parentheses refer to points in Figs. 4, 5, and 8.^bInitial particle size, 250–420 μm .^cParticle size reported to be 420–1680 μm .^dValved off.

Fig. 4. Consolidation of (250–420 μm) salt crystals in brine (see Table 2). Numbered points on the plots indicate a change in conditions. The curves were calculated from the expression in Table 1 with the following values for $\phi(0)$ and $Z(0)$: (a) 0.250 and 0.0389 cm, (b) 0.16 and 0.091 cm.

(350 μm) close to the original size (322 μm). In Run 202, considerable consolidation had occurred before measurements began, producing a smaller void fraction and requiring a much larger assumed particle size (0.09 cm) at the beginning of the measurements to account for the observed behavior. In this run, a systematic positive deviation of the data from the calculated curve appears as the void fraction falls below 0.05.

Machined specimens of commercially consolidated salt were compressed further, in two runs (203 and 204) in a column with a diameter of 9.54 cm and one run (205) in a column with a

Fig. 5. Further compaction of consolidated salt specimens in the presence of brine (see Table 2). (a) Hardy salt specimen; (b) Morton salt specimen; (c) Morton salt specimen; (d) halite specimen. Numbered points on the plots indicate a change in conditions. The curves were calculated from the expression in Table 1 with the following values for $\phi(0)$ and $Z(0)$: (a) 0.0727 and 0.18 cm, (b) 0.11 and 0.05 cm, (c) 0.085 and 0.07 cm, (d) 0.00189 and 0.12 cm.

diameter of 7.62 cm. In Run 206, a machined specimen of halite was similarly tested. As the curves for these cases (Fig. 5) indicate, after the initial data were fitted with the best choice of initial void fraction and particle size, the remaining consolidation proceeded more slowly than predicted. This is especially clear in Runs 205 and 206, in which constant applied stress and temperature were maintained throughout. Changes in applied stress and temperature, and especially the use of a temperature gradient in runs 203 and 204 make the projection of behavior from the previous isotherms runs more difficult. There seems to be little doubt that consolidation was generally less rapid than predicted at low void fractions. This may well have been caused by the decreasing permeability of the specimens, limiting the rate at which the brine could be excluded as consolidation proceeded. It is interesting to note, however, that under constant conditions the low void fraction still decreases in a nearly linear fashion with the logarithm of time.

3.1.2 Salt/Clay Consolidation

Five runs (302-306) were carried out on salt/clay mixtures to test the effects of applied stress, initial particle size, and clay content. The results of one other run (301, Table 3) were suspect because large volumes of saturated brine were passed in the early stages. This dissolved appreciable

Fig. 6. Consolidation of salt-clay mixtures in brine (see Table 3). The curves were calculated from the expression in Table 1 with the following values for $\phi(0)$ and $Z(0)$: (a) 0.375 and 0.0179 cm, (b) 0.39 and 0.0179 cm, (c) 0.354 and 0.0179 cm, (d) 0.34 and 0.0179 cm, (e) 0.282 and 0.052 cm.

amounts of salt by the pressure-solution effect and also may have caused some segregation. Accordingly, the results from Run 302 are likely to be more representative of the conditions being tested.

The most striking effect of clay is that, while it does not seem to appreciably affect the initial rate of consolidation, as consolidation proceeds the rate becomes greater than would be expected for pure salt (Fig. 6). In Run 302, with 1 wt % clay, this deviation begins at a void fraction of ~ 0.28 . In Run 303, with 2 wt % clay, the deviation begins at a void fraction of ~ 0.33 . In Run 304, again ~ 1 wt % clay but with a higher applied stress, the deviation is delayed until the void fraction reaches ~ 0.24 .

Table 3. Salt/clay consolidation tests (in 2.54-cm-diam column at $\sim 20^\circ\text{C}$)

Run no.	NaCl wt. (g)	Initial particle size (μm)	Clay content (wt %)	Applied stress (bar)	Elapsed time (min)	Brine passed (g)	Void Fraction	
							initial	final
301	78.2	150-177	1.018 ^a	31	8,920	2,967	0.427	0.157
302	76.4	150-177	0.999 ^a	31	30,500	1,595	0.385	0.003
303	76.2	150-177	1.991	31	20,000	46.1	0.387	0.046
304	76.1	150-177	0.900 ^a	93	22,800	123.6	0.381	0.011
305	75.1	250-420	0.571 ^a	93	23,100	124.8	0.418	0.041
306 ^b	755	420-600	0.284 ^a	155	40,500	139.2 ^c 81.3 ^d	0.392	0.046

^aEnough to coat each salt crystal with a layer $\sim 0.3 \mu\text{m}$ thick.

^bRun in the 7.62-cm-diam cell.

^cExcluded from ram end.

^dExcluded from anvil end.

In Runs 305 and 306, the particle size was increased and the amount of clay was adjusted to provide approximately the same film thickness as in the previous runs at 1 wt % clay ($\sim 0.3 \mu\text{m}$, assuming the crystal surfaces were coated uniformly). To fit the early consolidation in each of these runs, it was necessary to assume an abnormally large initial particle size. This might have been caused by some ordering of the crystals during the loading of the columns before consolidation began.

Though the rate was greater during the latter stages of consolidation with clay than it was for pure salt, the linear dependence on the logarithm of time was still exhibited. Consolidation was followed to a void fraction of less than 1% in two runs (302 and 304). Especially in these cases, as well as in two others, during the final stages of consolidation the void fraction exhibited a positive deviation from this logarithmic relationship. Again, this may be due to a permeability-limiting condition.

3.2 PERMEABILITY OF SALT/BRINE MIXTURES

From permeability measurements made during the consolidation of pure salt (Shor et al. 1981), it was found that within the considerable scatter of the data the permeability decreased as the sixth power of the decreasing void fraction during consolidation. The following expression was found to approximate the results, which included void fraction in the range 0.06 to 0.4:

$$\ln[Pm/Z(0)^2] \approx 21 + 6 \ln \phi(t), \quad (5)$$

where the permeability (Pm) is defined as the brine flow in g/min through an area of 1 cm^2 under a hydrostatic pressure gradient of 1 bar/cm (i.e., the units are $\text{g cm}^{-1} \text{ min}^{-1} \text{ bar}^{-1}$).

While the results (Fig. 7) show deviations from the line based on Eq. 5, generally they are not much beyond the scatter of the original data for pure salt-brine specimens. The results for Run 302

Fig. 7. Permeability of consolidating salt-clay mixtures (see Table 3). The dashed line, based on Eq. 5, has a slope of 6.

are systematically higher. Those for Runs 303 to 305 are systematically lower at higher void fractions, but at lower values the agreement improves. In Run 305, with ϕ below 0.1, there is some indication of a more rapid drop in permeability, but the data are generally too scattered to be certain. It appears that the presence of clay has no very pronounced effect on the permeability of consolidating salt at a given void fraction.

3.3 BRINE EXPULSION

One purpose in undertaking the study of large salt specimens was to investigate the migration of brine as a function of applied stress and temperature gradient. It soon became clear, however, that the flow of brine within the specimen was influenced more by consolidation of the specimen than by the temperature gradient. This is apparent from Run 202 (Fig. 8), in which the volume of brine expelled during consolidation matched fairly closely the decrease in volume of the specimen.

In Run 203, involving a machined specimen of Hardy salt, a low rate of water collection from flowing helium was observed in the early stages, indicating that brine expelled by consolidation was accumulating within the nickel frits at the ends of the specimen. Later, the amount of water collected began to catch up, and by the end of the run the total amount of brine excluded agreed satisfactorily with that originally taken up by the dry specimen and also with the volume loss from consolidation.

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Fig. 8. Expulsion of brine from salt specimens (see Table 2). (a) The last stages of consolidation of salt crystals; water was collected by flowing helium beginning at point 3; (b) Hardy salt specimen; helium flow was used throughout.

In Run 204, a machined specimen of Morton salt was predried and then exposed to upward flowing brine within the apparatus. After a stress of 1.5 bars was applied, the incoming flow of brine was shut off for a brief period. The amount of brine leaving the opposite end during this period (6.95 cm^3) was close to the volume loss occurring simultaneously from consolidation (6.41 cm^3). Later, when the incoming brine was again valved off, the amount of brine collected first at the opposite end and then at both ends was much less than the volume loss from consolidation. This suggests that brine had somehow been lost or, more likely, that a portion of the specimen contained gas.

In Run 205, involving a specimen of Morton salt that had been impregnated with brine under vacuum, the total volume of brine expelled matched the volume decrease quite closely [Fig. 9(e)]. Finally, in Run 206, a specimen of halite could be impregnated with only a small amount of brine initially, and this was nearly all expelled when the specimen was later stressed. The volume loss by consolidation, however, was about twice as great as the volume of brine expelled, suggesting that gas had not been removed completely before impregnating the specimen.

The fact that unequal amounts of brine typically were expelled from each end of a specimen was at first attributed to the effect of the temperature gradient or gravity. But there was also evidence that the specimens were not consolidating uniformly. This was provided by runs 205, 206, and 306, which were carried out in the cell equipped with a floating cylinder that was free to move as the salt compacted [Fig. 1 (a)]. The displacements of the ram and anvil pistons with respect to this cylinder were not equal [Fig. 9(c)] nor were they always uniform. Moreover, the amount of brine expelled from each end of the specimen did not always match the movement of the corresponding piston.

As a result of these complications, we were unable to learn anything definite about the effect of a temperature gradient. The only definite result of these measurements of brine loss or expulsion is that they correlate most strongly with consolidation. This would suggest that the trapping of brine is not an important mechanism of retention. Rather, it appears that flow of brine prevails throughout consolidation. This, of course, would be consistent with the nearly complete consolidation that occurs when salt specimens such as these are subjected to prolonged stress.

3.4 CRYSTAL INTERFACE PERMEABILITY

The results reported previously (Gilpartrick et al. 1982) showed that for a given temperature, axial stress (σ), and hydraulic pressure drop (ΔP), the radial flow of brine outward through the circular interface between two cylindrical salt crystals being pressed together decreased with time in accord with Eqs. 2 and 3. It was also tentatively suggested that the constant p_4 in these equations was proportional to the product $\sigma \Delta P^2$.

The more recent measurements at 20°C (Table 4) confirm the applicability of Eqs. 2 and 3 to the data, provided the permeability does not get too low (Fig. 10). The more recent data, however, indicate that p_4 is more nearly proportional to the product $\sigma \Delta P$ rather than $\sigma \Delta P^2$ (Fig. 11):

$$p_4(\text{bar/g}) \sim 0.01 \sigma \Delta P \quad (20^\circ\text{C}) , \quad (6)$$

where σ and ΔP are both expressed in bars. They also suggest that there is a limiting permeability $P(\infty)$ that depends on the hydraulic pressure drop (Fig. 12). While the results are scattered, it is

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Fig. 9. Expulsion of brine from Morton salt specimen (Run 205 in Table 2). (a) Brine volume vs time; (b) volume loss by consolidation vs time; (c) comparison of volume of brine expelled with volume loss by consolidation.

Table 4. Additional results on the permeability of salt-crystal interfaces to brine

Run no. ^a	Stress (bar)	Hydraulic pressure (bar)	Time (min)	Brine passed (g)	Pr(0) ^b (g min ⁻¹ bar ⁻¹)	P ₄ ^b (bar/g)	Pr(∞) ^b (g min ⁻¹ bar ⁻¹)	Standard deviation (g)
190-A2	5.26	0.06	5260	2.97	0.0120(5)	0.0116(16)		0.033
190-A3	5.26	0.33	1300	2.09	0.0066(1)	0.49(6)	0.0041(1)	0.008
190-B1	10.03	0.33	4610	3.62	0.00278(3)	0.029(2)		0.017
190-B2	10.03	0.73	4100	5.03	0.001736(4)	0.0091(7)		0.006
190-C1	36.7	1.14	5700	4.78	0.00101(2)	0.151(6)		0.029
190-C2	36.7	1.43	3080	2.28	5.45(4)E-4	0.067(9)		0.007
200-A1	62.5	1.35	5210	5.00	0.00143(2)	0.45(3)	1.9(5)E-4	0.018
210-A2	62.5	2.66	5700	6.09	8.7(4)E-4	0.55(3)		0.09
210-A4	56.3	1.38	11800	6.75	0.008(4)	0.72(3)		0.19
220-A2	15.6	0.75	2660	5.61	0.00437(4)	0.112(2)		0.019
220-B1	31.3	1.47	1460	0.70	7.9(3)E-4	11.4(4)	2.61(2)E-2	0.002
220-B2	31.3	3.1	4150	2.27	1.93(2)E-4	3.7(6)	1.69(1)E-4	0.003
220-C1	62.5	3.25	1401	1.10	5.36(4)E-4	14.5(2)	1.957(4)E-4	0.0008
220-D1	125	5.72	7220	2.24	6.45(11)E-5	8.25(12)	5.04(4)E-5	0.005
230-A1	50.0	2.7	5680	0.43	1.6(2)E-4	20.0(7)		0.01
240-A2	50.0	2.70	1020	0.124	8.9(5)E-5	63(7)	2.42(12)E-5	0.0005
250-A3	50.0	2.77	3250	0.300	5.7(1)E-5	11.5(3)		0.003

^aA new number was assigned when the crystal surfaces were renewed or reoriented.

^bPr(0), p₄, and Pr(∞) are parameters in Eqs. 28a and 29, which were used to fit the data. When a value for the limiting permeability Pr(∞) can not be extracted from the data, then Eq. 3 applies with P₃ = 1/Pr(0). The figures in parentheses indicate the uncertainty in the last digits.

^cStandard deviation of calculated from observed amount of brine collected.

suggested on the basis of a model to be presented in the next section that a more probable relationship is

$$Pr(\infty) \text{ (g min}^{-1} \text{ bar}^{-1}\text{)} \sim 0.0003/\Delta P^{1/2} \quad (20^\circ\text{C}) \quad (7)$$

4. DISCUSSION

The main purpose of this program has been to provide a better understanding of the consolidation and permeability of salt-brine mixtures. Such knowledge should be useful in assessing halite deposits as a storage medium for radioactive wastes. The expression (Eq. 1 and Table 1) proposed previously seems an adequate representation of the kinetics of salt consolidation in brine to void fractions as low as about 0.03. Further consolidation occurs more slowly than predicted by this expression, perhaps because the rate becomes limited by the permeability of the consolidating mass to the brine that must be excluded. To consider this possibility quantitatively, a model for the permeability of the interfacial region between salt crystals is needed.

Fig. 10. Time dependence of permeability for radial flow of brine in a crystal interface: (a) Run 190-C2; (b) Run 220-C1. Plotted points are running averages of five consecutive points.

Fig. 11. Dependence of parameter: p_4 (Eq. 3) on the applied stress σ and the hydraulic pressure drop ΔP .

We will present such a model, apply it to the results on the permeability of salt crystal interfaces, compare it to the results on the consolidation of polycrystalline salt in brine, and finally use it to make projections of the permeability of natural halite to brine.

4.1 A MODEL FOR CRYSTAL INTERFACE PERMEABILITY

The first system to be modeled is the one studied experimentally (Fig. 13), consisting of two cylindrical crystals of radius r_2 with right circular surfaces in contact and pressed together by an

Fig. 12. Observed limiting permeabilities for radial flow of brine in a crystal interface (see Table 4). The line indicates the dependence on ΔP predicted by model described in Sect. 4.1.

axial stress σ . There is an axial hole of radius r_1 in the upper crystal, that defines the inner boundary of the interface and through which brine is forced into the interface under a hydraulic pressure $P(r_1)$. The inlet salt concentration $c(r_1)$ equals the saturation value for NaCl at ordinary pressure and a controlled temperature.

For modelling purposes we will imagine the liquid film between the crystals to be continuous, providing a medium in which salt may be dissolved by the pressure solution effect and carried off. [The pressure solution effect refers to the increase in solubility of salt that results from an increase in pressure; its magnitude for NaCl has been reviewed by Jenks (1979).] We will assume the salt exposed to the brine is under a pressure arising from the applied stress σ , while the liquid film is subject only to the hydraulic pressure drop (dP/dr) causing flow. The points of contact between the two surfaces that maintain their separation will be ignored for the moment, and the surfaces will be assumed to be smooth. Such a representation was employed by Weyl (1959) in a somewhat similar model.

4.1.1 Steady-State Case

We will consider first the consequences of a steady-state condition at a given applied stress and hydraulic pressure drop. Because of pressure solution, salt will be dissolved from both adjacent sur-

faces as the brine passes radially outward. The average concentration of the solution at a distance r from the axis, $c(r)$ in moles per unit volume, will increase such that

$$\{c(r) - c(r_1)\}Q = \pi(r^2 - r_1^2)R\rho_s/M, \quad (8)$$

where

M = the molecular weight of NaCl,

ρ_s = its density,

R = the rate at which the overall axial length of the crystal pair is decreasing,

Q = the volume flow rate of brine.

Because R and Q are independent of r , this equation shows that c must increase linearly with r^2 .

For the crystal surfaces to remain in good contact as time passes, it is necessary that the net dissolution rate be the same at all locations in the interface. If this dissolution process is assumed to be diffusion limited and to take place equally at both surfaces, then it follows that the flux normal to the surface will similarly be a constant and (from Eq. 8) equal to

$$J = \frac{\{c(r) - c(r_1)\}Q}{2\pi(r^2 - r_1^2)} = \frac{R\rho_s}{2M}. \quad (9)$$

From Fick's First Law, the concentration gradient at the surface must also be a constant:

$$\left(\frac{\partial c}{\partial z}\right)_{z=h} = \frac{R\rho_s}{2MD}, \quad (10)$$

where D is the diffusion coefficient. Here we have chosen the origin of the z direction as the mid-point between the crystal surfaces and defined the separation distance at r to be $2h$ (Fig. 13).

The following integral equation relates the concentration $c(r,z)$ at a particular location r,z in the liquid film to the average concentration $c(r)$, which appears in Eqs. 8 to 10.

$$c(r) \cdot Q = 4\pi r \int_0^{h(r)} c(r,z) \cdot u(r,z) dz. \quad (11)$$

Here $u(r,z)$ is the fluid velocity at the position r,z , which must also satisfy the integral equation

$$Q = 4\pi r \int_0^{h(r)} u(r,z) dz. \quad (12)$$

The functions $c(r,z)$ and $u(r,z)$ should satisfy the appropriate equations for diffusion and fluid flow as well as Eqs. 10, 11, and 12. Fortunately, such rigor is unwarranted because the results obtained are not sensitive to the choice of these functions. Two were tested for each (Table 5)—functions linear and parabolic in z for $c(r,z)$ that satisfy Eq. 10 and functions independent of and parabolic in z for $u(r,z)$ that satisfy Eq. 12. When the four possible combinations of these functions were introduced into Eq. 11, the expressions for $c(r)$ obtained on integration were all of the same form:

$$c(r) = c_s - a \left(\frac{R\rho_s}{MD} \right) h(r), \quad (13)$$

Fig. 13. Schematic representation of a circular crystal interface under stress with radial brine flow under steady-state conditions. The interfacial distance $2h$ should decrease linearly as the square of the radial distance r .

differing only in the numerical coefficient a (Table 5). In this equation c_s is the saturation solubility at the salt-brine interface. The small variation in the value of a (from 0.167-0.313) suggests that more rigorous choices for the functions $c(r,z)$ and $u(r,z)$ are indeed unwarranted considering the crudeness of this model. We will adopt the most realistic case tested, that in which parabolic functions are assumed for $c(r,z)$ and $u(r,z)$, giving

$$a = 0.2 . \quad (13a)$$

Eliminating $c(r)$ values between Eqs. 8 and 13,

$$h(r) = h(r_2) + \frac{\pi D(1 - \alpha)}{aQ} (r_2^2 - r^2) , \quad (14)$$

where

$$\alpha = \frac{c_s(r_2) - c_s(r)}{c(r_2) - c(r)} . \quad (14a)$$

Table 5. The effect of assumed concentration and velocity functions

Brine concentration

Linear: $c(r,z) = c_s - \frac{R\rho_s}{2MD}(h - z)$

Parabolic: $c(r,z) = c_s - \frac{R\rho_s}{4MD}\left[\frac{h^2 - z^2}{h}\right]$

Brine velocity

Constant: $u(r,z) = \frac{Q}{4\pi rh}$

Parabolic: $u(r,z) = \frac{3Q}{8\pi r}\left[\frac{h^2 - z^2}{h^3}\right]$

Resulting expression for $c(r)$

$$c(r) = c_s - a\left[\frac{R\rho_s}{MD}\right]h$$

Case	$c(r,z)$	$u(r,z)$	a
1	Linear	Constant	1/4
2	Linear	Parabolic	5/16
3	Parabolic	Constant	1/6
4	Parabolic	Parabolic	1/5

The simplest assumption that could be made is that the saturation value at the salt-brine interface, c_s , is independent of r . Then alpha vanishes and we obtain a simple expression predicting an increase in the film half thickness h as r^2 decreases, the rate depending on the diffusion coefficient and the brine flow rate. Unfortunately, for small flow rates the calculated increase in h becomes impossibly large. For example, at a flow rate of 10^{-6} cm³/s (about the minimum found in our measurements) and with a diffusion coefficient of 1.45×10^{-5} cm²/s, h would increase by 73 cm from its minimum value at $r_2 = 0.57$ cm to its maximum value at $r_1 = 0.08$ cm! Hence it appears that at least one of the assumptions made thus far—smooth walls, diffusion-limited salt dissolution, and a saturation-solubility (c_s) independent of r —must be an unacceptable approximation.

Without a detailed knowledge of the conditions within the interface, it is not possible to decide how to improve upon Eq. 14. We will assume therefore that the form is correct and investigate the consequences of two possible limiting conditions:

1. The half thickness of the interfacial film is independent of r . This could occur if the value of c_s were to change with r at the same rate as the average concentration $c(r)$; alpha then goes to unity and h becomes independent of r . Alternatively, the same result is of course obtained if the effective diffusion coefficient D goes to zero.

2. The half thickness of the interfacial film increases from its minimum value, $h(r_2)$, at the outer periphery to its maximum value, $h(r_1)$, at the inner periphery by an amount much greater than its minimum value; that is,

$$[h(r_1) - h(r_2)]/h(r_2) \gg 1 .$$

This could be the case if the saturation value for brine at the interface, c_s , is independent of r (α is zero) and the effective diffusion coefficient D is for some reason much smaller than the measured value for brine. For $h(r_1)$ values to remain acceptably small (e.g., <0.01 cm), D would have to be less than 10^{-9} cm²/s. Such a small value would be conceivable if salt dissolves from very localized sites and reaches the flowing brine by a tortuous path.

We now proceed to derive an expression for the flow rate Q as a function of h and the hydraulic pressure drop. From the treatment of Bird et al. (1960, p. 106) for the present radial geometry, it can be shown that

$$Q = - \frac{4\pi r}{3\eta} \left(\frac{dP}{dr} \right) h(r)^3 , \quad (15)$$

where η is the viscosity. Rearranging and integrating from r_1 to r_2 ,

$$\Delta P = \frac{3\eta Q}{4\pi} \int_{r_1}^{r_2} \frac{dr}{rh^3} , \quad (16)$$

where $\Delta P = P(r_1) - P(r_2)$. After insertion of the expression for $h(r)$ given by Eq. 14 with α independent of r , the integral on the right reduces to a standard form, giving

$$\Delta P = \frac{3\eta Q}{8\pi h(0)^3} [I]_{r_1}^{r_2} , \quad (17)$$

where $h(0)$ is the half thickness the interfacial film would reach if the inner radius of the interface could go to zero:

$$h(0) = h(r_2) + Br_2^2 , \quad (17a)$$

$$B = \frac{\pi D(1 - \alpha)}{aQ} , \quad (17b)$$

$$I = \frac{h(0)}{h(r)} + \frac{h(0)^2}{2h(r)^2} + \ln \frac{r^2}{h(r)} . \quad (17c)$$

If h is independent of r , then Eq. 17 simplifies and may be rearranged to

$$Q = C_r' \Delta P h^3 \quad (h \text{ independent of } r) , \quad (18)$$

where

$$C_r' = \frac{4\pi}{3\eta \ln(r_2/r_1)} . \quad (18a)$$

If $h(0)/h(r)$ is much larger than unity, then

$$h(0) \rightarrow Br_2^2, [I]_{r_1}^{r_2} \rightarrow \frac{B^2 r_2^4}{2h(r_2)^2}$$

and Eq. 17 after rearrangement becomes

$$Q = C_r'' \Delta P^h h(r_2) \quad (h \text{ decreases as } r^2 \text{ increases}), \quad (19)$$

where

$$C_r'' = \left(\frac{80D(1 - \alpha)}{3\eta} \right)^h \pi r_2. \quad (19a)$$

Thus if h is constant from r_1 to r_2 , then the steady-state flow rate is proportional to the hydraulic pressure drop and to the cube of h (Eq. 18). This is the classic result for radial flow between plane-parallel plates. If, however, h decreases linearly as r^2 increases and the change is sufficiently large, the flow becomes proportional to the square root of the hydraulic pressure drop and to the first power of the minimum value of h (Eq. 19).

4.1.2 Time-Dependent Case

While we have assumed in the steady-state case that salt is being dissolved continuously at all points on both surfaces in such a way as to maintain the distance of separation ($2h$) independent of time, it is reasonable to suppose that the irregularities that maintain the separation between the crystal surfaces will in fact dissolve preferentially and cause the separation distance to decrease with time. While the details of the process are not known, the following assumed rate expression for h produces a final result consistent with the present experimental data:

$$\frac{dh}{dt} = -kR[h(t) - h(\infty)]. \quad (20)$$

Here $h(\infty)$ is the lower limit of h , corresponding to the limiting smoothness a dissolving crystal surface can attain; R is the rate at which salt is being dissolved uniformly from both surfaces and is given by Eq. 8 as

$$R = \frac{[c(r_2) - c(r_1)]QM}{\pi(r_2^2 - r_1^2)\rho_s}. \quad (21)$$

We may make the approximation that the increase in salt concentration $[c(r_2) - c(r_1)]$ is proportional to the applied stress σ (Jenks 1979):

$$c(r_2) - c(r_1) = K_\sigma \sigma. \quad (22)$$

The rate expression for h thus becomes

$$\frac{dh}{dt} = -kN_\sigma Q[h(t) - h(\infty)], \quad (23)$$

where

$$N = \frac{K_s M}{\pi(r_2^2 - r_1^2)\rho_s} \quad (23a)$$

We further assume that the rate of change of h with time is slow enough that the stationary state condition leading to Eqs. 18 or 19 is not appreciably upset.

Considering first the case in which h is independent of r , we integrate Eq. 23 using Eq. 18 and with σ and ΔP independent of time to obtain

$$\left[\frac{1}{2h(t)^2 h(\infty)} + \frac{1}{h(t)h(\infty)^2} + \frac{\ln[1 - h(\infty)/h(t)]}{h(\infty)^3} \right]_{h(0)}^{h(t)} = -kNC'_{,\sigma} Q \Delta P t \quad (24)$$

When $h(t) \gg h(\infty)$ and after replacement of h by use of Eq. 18, this simplifies to*

$$\frac{1}{Q(t)} = \frac{1}{Q(0)} + 3kN\sigma t \quad (25)$$

The limiting flow reached after sufficient time is given by Eq. 18 if $h(\infty)$ is specified.

Considering next the case in which the film thickness changes with r , we integrate Eq. 23 with the use of Eq. 19 to obtain.

$$\frac{1 - Q(\infty)/Q(t)}{1 - Q(\infty)/Q(0)} = \exp[-kN\sigma Q(\infty)t] \quad (26)$$

For small values of the exponential, that is, at sufficiently small values of the time t , this reduces to

$$\frac{1}{Q(t)} = \frac{1}{Q(0)} + kN\sigma t \quad (27)$$

Equations 25 and 27 can be placed in the same form as Eq. 3, with

$$Pr = Q\rho_b/\Delta P \quad (28a)$$

and giving

$$p_3 = 1/Pr(0) \quad (28b)$$

and

$$p_4 = \frac{\sigma}{\rho_b} \Delta P \quad (h \text{ independent of } r) \quad (28c)$$

or

$$p_4 = \frac{kN}{\rho_b} \sigma \Delta P \quad (h \text{ decreases as } r^2 \text{ increases}) \quad (28d)$$

*Equation 25 is obtained by replacing $\ln(1-x)$ with $-x - x^2/2 - x^3/3$ [$x = h(\infty)/h(t)$] in Eq. 24.

The general expression for Pr in the latter case is

$$\frac{1 - Pr(\infty)/Pr(t)}{1 - Pr(\infty)/Pr(0)} = \exp\left[-\left(\frac{kN}{\rho_b}\right)Pr(\infty)\sigma \Delta P t\right], \quad (29)$$

and for the limiting permeability, we have

$$Pr(\infty) = C_r' \rho_b h(\infty)^3 \quad (h \text{ independent of } r) \quad (30)$$

or

$$Pr(\infty) = C_r'' \rho_b \frac{h(r_2, \infty)}{\Delta P^{1/2}} \quad (h \text{ decreases as } r^2 \text{ increases}). \quad (31)$$

Thus, while the two assumptions about h give the same time dependence for relatively large values of Pr , this is not true of the limiting permeability. For constant h , $Pr(\infty)$ should not depend on ΔP , while if h decreases substantially in the direction of radial flow, $Pr(\infty)$ should vary inversely as the square root of ΔP .

4.2 COMPARISON WITH OBSERVED INTERFACE PERMEABILITIES

The limiting permeabilities observed for crystal interfaces (Fig. 12), as represented by Eq. 7, may be compared with Eqs. 30 and 31 after introducing the units employed in the measurements (flow rates in g brine/min, ΔP and σ in bars) and the following parameter values:

$$r_1 = 0.0795 \text{ cm},$$

$$r_2 = 0.57 \text{ cm},$$

$$\eta = 0.0199 \text{ g cm}^{-1} \text{ s}^{-1} \quad [\text{estimated from International Critical Tables (ICT)}], \text{ and}$$

$$\rho_b = 1.2011 \text{ g/cm}^3 \quad (\text{ICT}).$$

If h is independent of r , then the limiting permeability should be given by

$$Pr(\infty) (\text{g min}^{-1} \text{ bar}^{-1}) = 7.7 \cdot 10^9 h(\infty)^3 \quad (32)$$

(with h in cm). Because the limiting permeabilities were of the order of $10^{-4} \text{ g min}^{-1} \text{ bar}^{-1}$, this suggests minimum values for h of about $0.2 \mu\text{m}$.

If h decreases sufficiently as r increases, then Eq. 31 applies, giving

$$Pr(\infty) (\text{g min}^{-1} \text{ bar}^{-1}) = 4.72 \cdot 10^6 D^{1/2} (1 - \alpha)^{1/2} h(r_2, \infty) / \Delta P^{1/2}. \quad (33)$$

By comparison with Eq. 7, we find the limiting value of the half thickness of the interface at the periphery to be

$$h(r_2, \infty) (\text{cm}) = \frac{6.4 \times 10^{-11}}{D^{1/2} (1 - \alpha)^{1/2}}. \quad (34)$$

As we have already noted, in this case the product $D(1 - \alpha)$ must be less than $10^{-9} \text{ cm}^2/\text{s}$. Hence we may conclude in this case that the limiting value of $h(r_2)$ is $0.021 \mu\text{m}$ or greater.

Next, we estimate the rate constant k as follows: The numerical factor N (Eq. 23a) is evaluated with

$$K_s = 1.58 \times 10^{-6} \text{ mol cm}^{-3} \text{ bar}^{-1} \text{ (after Jenks, 1979),}$$

$$\rho_s = 2.1631 \text{ g cm}^{-3}, \text{ and}$$

$$\pi (r_2^2 - r_1^2) = 1.001 \text{ cm}^2.$$

giving

$$N = 4.265 \times 10^{-5} \text{ cm}^{-2} \text{ bar}^{-1}. \quad (35)$$

Comparing corresponding coefficients of t in Eqs. 6, 28a, and 28b,

$$k(\text{cm}^{-1}) = 100 \quad (h \text{ independent of } r), \quad (36a)$$

$$k(\text{cm}^{-1}) = 300 \quad (h \text{ decreases as } r^2 \text{ increases}). \quad (36b)$$

4.3 THE PERMEABILITY OF POLYCRYSTALLINE SALT TO BRINE

We will now apply this model to estimate the permeability of a planar interface between salt crystals with unidirectional flow (Fig. 14) and then the permeability of idealized polycrystalline aggregate (Fig. 15).

Fig. 14. Schematic representation of a linear crystal interface under stress with unidirectional brine flow under steady-state conditions. The interfacial distance $2h$ should decrease linearly with the distance y .

Fig. 15. Idealized representation of brine flow in consolidated salt.

4.3.1 Unidirectional Flow in a Crystal Interface

The development for this case parallels that for the radial case. Referring to Fig. 14, the width of the interface is w , the length in the direction of flow is l , and the coordinate in this direction is labelled y . Equation 8 then becomes

$$[c(y) - c(0)]Q = wyR\rho_s/M \quad (37)$$

The constant concentration gradient normal to the surface (Eq. 10) is now

$$\left(\frac{\partial c(y,z)}{\partial z} \right)_{z=h} = \frac{[c(y) - c(0)]Q}{2wyD} = \frac{R\rho_s}{2MD} \quad (38)$$

The integral equation relating $c(y,z)$ to $c(y)$ (compare Eq. 11) is

$$c(y)Q = 2w \int_0^h c(y,z)u(y,z) dz \quad (39)$$

and that relating Q to $u(y,z)$ is

$$Q = 2w \int_0^h u(y,z) dz \quad (40)$$

When parabolic functions analogous to those in Table 5 are used for the concentration $c(y,z)$ and the velocity $u(y,z)$ across the interfacial film, the results are

$$h(y) = h(l) + \frac{wD(1-\alpha)}{aQ}(l-y) \quad (41)$$

$$Q = \frac{2w}{3\eta} \left(\frac{dP}{dy} \right) h(y)^3. \quad (42)$$

If we assume that h is independent of y , then the expression for the flow again takes the form of Eq. 18 but with the constant C_r' replaced by

$$C_u' = \frac{2w}{3\eta l}. \quad (43)$$

Finally, if h decreases as y increases by an amount that is large compared to its minimum value at $y = l$, then the same form as Eq. 19 is obtained but with the constant C_r'' replaced by

$$C_u'' = \left[\frac{20D(1 - \alpha)}{3\eta} \right]^{\frac{1}{n}} w. \quad (44)$$

Thus the flow rate exhibits the same dependence on the hydraulic pressure drop and on h as it did for radial flow.

4.3.2 Unidirectional Flow Through Polycrystalline Salt

These expressions for unidirectional flow may be applied to the configuration shown in Fig. 15 when we note that the number of interfaces running in the direction of flow is $2w/Z$ (Z being the crystal size). Because the width of each such interface is w , then the aggregate width of all interfaces in the direction of flow is $2w^2/Z$. Hence the total flow through the aggregate should be

$$Q = C_u' \frac{2w}{Z} \Delta P h^3 \quad (h \text{ independent of } y), \quad (45)$$

$$Q = C_u'' \frac{2w}{Z} \Delta P^{\frac{1}{n}} h(l) \quad (h \text{ decreases as } y \text{ increases}), \quad (46)$$

where C_u' and C_u'' are given by Eqs. 43 and 44.

If the permeability of the configuration in Fig. 15 is defined as the mass flow of brine per unit area per unit hydraulic pressure gradient, then

$$Pm = \frac{Q/\rho_b}{w^2 \Delta P}. \quad (47)$$

If h is independent of y , Eqs. 18, 28a, 43, 45, and 47 may be combined to relate Pm to the permeability, Pr , measured for radial flow:

$$Pm = \frac{2 l C_u' \rho_b h^3}{Z w},$$

$$Pm = \frac{\ln(r_2/r_1)}{\pi Z} Pr = \frac{0.627}{Z} Pr \quad (h \text{ independent of } r \text{ or } y). \quad (48)$$

If h decreases in the direction of flow, Eqs. 19, 28a, 44, 46, and 47 may be used to show that

$$Pm = \frac{1}{\pi r^2 Z} Pr = \frac{0.558l}{Z} Pr \quad (h \text{ decreases as } r \text{ or } y \text{ increases}) . \quad (49)$$

Because in our experiments the height of consolidating columns was typically a few centimeters, these two expressions (Eqs. 48 and 49) give about the same predicted values for $Pm(\infty)$ from the observed limiting values of Pr . With Pr of the order of $10^{-4} \text{ g min}^{-1} \text{ bar}^{-1}$, the limiting permeability thus predicted in salt consolidation is about $10^{-4}/Z \text{ g min}^{-1} \text{ bar}^{-1}$. The particle size Z in our measurements was usually in the range 0.01 to 0.04 cm, so $Pm(\infty)$ should have been in the range 0.003 to 0.01. This produces predicted values of $Pm(\infty)/Z^2$ in the range of 1 to 100, near the lower limit of the measured values plotted in Fig. 7. Thus the limiting brine flow rates found in the crystal interface measurements are, within the substantial scatter involved, consistent with permeabilities measured during the consolidation of polycrystalline salt.

It is more difficult to predict when the permeability might become the limiting factor in the final stages of consolidation. A crude estimate may be made as follows: If the brine exclusion rate limits consolidation, then the hydraulic pressure drop producing the exclusion should be comparable to the uniaxial stress divided by half the length of the column. Hence, for a consolidating column of length l and radius r ,

$$Q \sim \frac{2\pi r^2 \sigma}{l \rho_b} Pm(\infty) ,$$

$$\frac{d\phi}{dt} \sim -\frac{2\sigma}{l^2 \rho_b} Pm(\infty) .$$

Introducing the two expressions for $Pm(\infty)$ in terms of Pr (Eqs. 48 and 49),

$$\frac{d\phi}{dt} \sim -\frac{1.0\sigma}{Zl^2} Pr(\infty) \quad [h(\infty) \text{ constant}],$$

$$\frac{d\phi}{dt} \sim -\frac{0.93\sigma}{Zl} Pr(\infty) \quad [h(\infty) \text{ decreases in the direction of flow}].$$

Inserting typical values of $\sigma \approx 50$ bars, $Z = 0.02$ cm, $l = 10$ cm, and $Pr = 0.0001 \text{ cm}^3 \text{ min bar}$, the rate of decrease in the void fraction in the limit should be 0.0025 min^{-1} if $h(\infty)$ is constant and about ten times larger if $h(\infty)$ decreases in the direction of flow. These values are relatively large, suggesting that permeability remains too high to account for the slower rates of decrease in the void fraction observed in the latter stages of consolidation.

Table 6 lists permeabilities that are predicted by Eqs. 48 and 49 for conditions that might be found in a salt repository. For the case that $h(\infty)$ is constant, $Pr(\infty)$ was again taken to be $0.0001 \text{ g min}^{-1} \text{ bar}^{-1}$. For the case of a variable h , Eq. 6 was used to evaluate $Pr(\infty)$.

Table 6. Predicted limiting permeability of polycrystalline salt

Particle size	Permeability, ^a cm ² min ⁻¹ bar ⁻¹ (μDarcy)	Brine flow ^b (μm min ⁻¹)		
		ΔP (bar)		
		1	10	100
0.1	5 × 10 ⁻⁴ 17	14	44	140
1	5 × 10 ⁻⁵ (1.7)	1.4	4.4	14
10	5 × 10 ⁻⁶ (0.17)	0.14	0.44	1.4

^aCalculated from Eq. 48 with $Pr(\infty) = 0.0001$ (from Fig. 12), based on the assumption that $h(\infty)$ is a constant.

^bCalculated from Eq. 49 with $Pr(\infty)$ given by Eq. 7 (from Fig. 12), based on the assumption that $h(\infty)$ decreases in the direction of flow. The units are 10⁻⁴ cm³ of brine flow per min per cm² of surface normal to the direction of flow. In this case, the flow rate is proportional to the square root of the hydraulic pressure drop.

5. SUMMARY AND CONCLUSIONS

1. Additional results on the kinetics of consolidation of sized salt crystals in brine (Fig. 4) are consistent with the previously reported rate expression (Eq. 1 and Table 1). The further consolidation of salt-block specimens impregnated with brine is found to be slower than predicted (Fig. 5).

2. The consolidation of salt/clay mixtures in brine proceeds more rapidly than predicted (Fig. 6) and was followed almost to completion. Their permeability to brine, however, was similar in its dependence on the void fraction to that observed previously for pure salt (Fig. 7).

3. Studies of the migration of brine within stressed salt specimens indicated that the most important effect was that of brine exclusion as consolidation proceeds (Figs. 8 and 9). There was no evidence that significant amounts of brine were trapped. Also, no clear effect of a temperature gradient on brine migration could be discerned.

4. In studies of the permeability of salt crystal interfaces to brine, the previously reported linear increase of the reciprocal permeability on the time (Eq. 3) was confirmed (Fig. 10), though the rate of increase appears to be more nearly proportional to the product of the applied stress and the hydraulic pressure drop, $\sigma\Delta P$, rather than to $\sigma\Delta P^2$ as previously suggested (Eq. 6 and Fig. 11). There is also evidence in these measurements that a limiting permeability is reached that decreases with increasing hydraulic pressure drop (Eq. 7 and Fig. 12).

5. A model has been developed to rationalize the observed permeability behavior of stressed salt-crystal interfaces (Fig. 13). If it is assumed that the rate of salt removal from the contacting

surfaces (as a result of the pressure-solution effect) is diffusion limited, for smooth surfaces it can be shown that for the radial flow geometry studied the interfacial thickness will decrease linearly with the square of the increasing radial distance r (Eq. 14). However, the effective diffusion coefficient required is much (at least 10,000 times) smaller than the expected value for brine. Two limiting steady-state cases were tested in which (1) the film half thickness is independent of r and (2) it decreases substantially as r^2 increases. The former predicts that the brine flow rate will be proportional to $\Delta P h^3$ (Eq. 18), while the latter predicts that the flow will be proportional to $\Delta P^{1/2} h(r_2)$ (Eq. 19, where $h(r_2)$ is the (minimum) film half thickness at the outer boundary of the circular interface.

6. An equation is suggested (Eq. 20) for the decrease in the film half thickness h with time that is reasonably consistent with the observed time dependence of the permeability (Eqs. 6 and 7 and Figs. 11 and 12).

7. This model is extended to the case of unidirectional flow in a crystal interface (Fig. 14) and then to polycrystalline salt (Fig. 15). The permeability behavior of polycrystalline salt may be predicted for the two limiting cases of the model from the measurements made with radial geometry (Eqs. 48 and 49). From this evidence it does not appear that the observed salt consolidation rates were limited by permeability. The behavior of aggregates of larger salt crystals is also suggested for the two limiting cases of the model (Table 6).

The most important conclusion to be drawn from this work is that, under stress, polycrystalline salt consolidates readily in the presence of brine with fairly predictable kinetics and with the exclusion of nearly all the brine. The presence of clay seems to hasten this process, possibly through a lubricating action that eases the reorientation of crystals. Because it does not greatly affect the permeability, the clay evidently tends to collect in void spaces rather than in the crystal interfaces.

The rate expression for the decrease in the interfacial film thickness (Eq. 20) that accounts for the time dependence of the permeability (Eq. 6) is a reasonable one and suggests that the model developed here may be a useful predictive tool. Unfortunately, the results for the limiting permeability are not reproducible enough (Fig. 12) to permit a choice between the limiting forms of the model. If the dependence on the square root of the hydraulic pressure drop suggested by Eq. 7 is correct, then the film thickness decreases in the direction of flow and Darcy flow cannot be assumed. Both the magnitude of the limiting permeabilities and their dependence on the hydraulic pressure drop should be verified by additional measurements.

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GLOSSARY OF SYMBOLS

- a = A numerical constant (Eqs. 13, 14, 41).
 $\bar{c}(r)$ = Average brine concentration (mole/volume) at radial distance r .
 $c(r,z)$ = Actual brine concentration at radial distance r and distance z from the midplane of the interfacial film.
 c_s = saturation concentration of brine when salt is under stress σ .
 C_r', C_r'' = Constants (Eqs. 18a and 19a) for radial flow.
 C_u', C_u'' = Constants (Eqs. 43 and 44) for unidirectional flow.
 D = Diffusion coefficient of NaCl in brine.
 η = Brine viscosity.
 G = Flow rate of brine (mass/time).
 $h(r)$ = Half thickness of interface at radial distance r .
 $h(r_2, \infty)$ = Minimum half thickness at outer perimeter (radial flow).
 $h(l, \infty)$ = Minimum half thickness at exit (unidirectional flow).
 J = Flux of dissolving NaCl.
 k = A rate constant (Eqs. 20 and 36).
 K_s = Constant relating NaCl solubility to stress on the solid (Eq. 22).
 l = Extent of an interface in the direction of (unidirectional) flow.
 M = Molecular weight of NaCl.
 N = A numerical constant (Eq. 23a).
 $p_1 - p_4$ = Adjustable parameters in Eqs. 1 to 3.
 ΔP = Hydraulic pressure drop.
 $P(r)$ = Hydraulic pressure at radial distance r .
 Pm = Permeability to brine flow (mass flow rate per unit area per unit hydraulic pressure gradient).
 Pr = Permeability to brine flow for radial geometry (mass flow rate per unit hydraulic pressure drop).
 ϕ = Void fraction in a salt-brine specimen.
 Q = Flow rate of brine (volume/time).
 ρ_s = Density of crystalline NaCl.
 ρ_b = Density of brine.
 r = Coordinate in radial direction.
 r_1, r_2 = Inner and outer radius of circular interface.
 R = Rate at which salt is dissolved in interface (length/time).
 t = Time.
 $u(r,z)$ = Fluid velocity at position r, z in the interfacial fluid.
 w = Extent of an interface normal to unidirectional flow.
 y = Coordinate in the direction of (unidirectional) flow.
 z = Coordinate normal to the midplane of the interface.
 Z = Crystal size.

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