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THE EFFECT OF PRESSURE AND TEMPERATURE ON THE THERMAL PROPERTIES OF A SALT AND A QUARTZ MONZONITE

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ABSTRACT

Measurements have been made of thermal conductivity, diffusivity, and linear expansion as a *function of temperature* (to 573 K) and hydrostatic pressure (to 50 MPa) on two rocks, Avery Island domal salt and Climax Stock quartz monzonite. The purpose of the measurements has been to determine rock thermal properties at in situ conditions of an underground nuclear waste repository and to understand the way in which existing *microfractures and thermally induced fresh microfractures* affect the thermal properties. For Avery Island salt the thermal properties do not show any pressure dependence and are approximately the same values as for single crystal halite at 0.1 MPa. The lack of pressure dependence is attributed to the [illegible] and to its low [illegible]. For Climax Stock quartz monzonite no pressure dependence of thermal diffusivity has been resolved, but conductivity does show a drop of approximately 10% with decreasing pressure from 50 to 3 MPa. The pressure dependence is not measurably altered by heating the rock to as high as 473 K under 50 MPa. Our measurements so far on the thermal conductivity of quartz monzonite vs. temperature and pressure are in *agreement with predictions of the* Walsh and Decker (1966) model of thermal conductivity vs. crack porosity based on independent measurements of crack porosity vs. temperature and pressure for the same quartz monzonite. We therefore believe that heating *to temperatures greater than 473 K at 50 MPa*, or heating to lower temperatures at lower pressures will [illegible] reduce thermal conductivity of the quartz monzonite.

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INTRODUCTION

Prediction of the long term thermomechanical response of an underground repository to thermal loading from nuclear waste requires knowledge of rock properties in situ and an understanding of the mechanism by which these properties are changed by thermal loading. Accurate prediction is necessary because of the [illegible] heat and radioactivity [illegible] to be breached and because of the [illegible] of performing long term [illegible] this possibility.

Most past measurements of thermal properties of rocks have been made under atmospheric pressure, but [illegible] waste [illegible] experimental [illegible] room pressure [illegible] to the underground repository situation [illegible] reasons [illegible] most rocks at the [illegible] porosity generally impedes thermal [illegible], and since static pressure acts to [illegible] pressure can be expected to affect thermal properties. The relationship between crack porosity and thermal conductivity has been studied [illegible] and experimentally (see Sweet, 1979, [illegible] constructed a sample that [illegible] the arrangement of cracks in a [illegible] model with [illegible] rocks. [illegible] that the application of pressure to squeeze out a crack porosity of [illegible] in a [illegible] rock will cause a [illegible] increase in thermal conductivity.

The second reason for pressure measurements [illegible] at pressure is that heating a rock, uniformly or otherwise, can cause crack porosity to increase because of local stresses due to mismatches [illegible] thermal expansivities (Richter and Simmons, 1974, Cooper and Simmons, 1977, Johnson et al., 1978, [illegible] and Gangi, 1980). Confining pressure acts to reduce the growth of crack porosity (Wong and Brace, 1979, Heard, 1980; Page and Heard, 1981, Bauer and Handin, 1983), and, by the discussion above, would push thermal conductivity and diffusivity above room pressure values. We report here measurements of thermal conductivity, diffusivity, and linear expansion carried out in an apparatus designed to make such measurements on large grained rocks at physical and chemical conditions expected in an underground waste repository. Data are reported for the first two rock types tests: a domal salt and a quartz monzonite.

SAMPLE MATERIAL

The salt samples were cored from a 400-mm diameter core (C-11), supplied by RE/SPEC Inc., Rapid City, South Dakota. The core was taken from the floor of workings at the 165-m level at the site of an in situ heater test in the Avery Island domal salt formation. Chemistry of samples from the Avery Island Mine show roughly 99.1% NaCl (by weight), 0.[illegible]% water insolubles, 0.2% $CaSO_4$ (anhydrite) and 0.2% water (Kaufmann, 1960). The rock salt has a porosity of approximately 1% and the grain size of the halite ranges from 2.5 to 15 mm with an average of 7.5 mm (Carter and Hansen, 1980). The density of the rock salt has not been measured. The density of pure NaCl at room temperature is 2.16 Mg m^{-3}.

The Climax Stock is located at the Nevada Test Site, Nevada. The quartz monzonite from the stock has been characterized in detail by Izett (1960) and Maldonado (1977). Average composition of several samples taken from the U-15a shaft [illegible] distant from the spent fuel repository was determined as 28% by weight quartz, 25% alkali feldspar, 40% plagioclase, 5% biotite, and 1% accessory minerals (Maldonado, 1977). Grain size in the matrix is 1-2 mm but is larger in quartz phenocrysts (5-10% by volume) averaging 4 mm in diameter and in large orthoclase phenocrysts [illegible] averaging [illegible] mm in length with some as long as 150 mm. The orthoclase phenocrysts are uniformly distributed in the rock. A volume of rock 40 cm^{3} or more which does not encounter at [illegible] phenocryst [illegible]. The rock has a [illegible] porosity of approximately [illegible] (Page and Heard, [illegible]) and has a density of approximately 2.6[illegible] Mg m^{-3}.

EXPERIMENTAL [illegible]

[illegible] apparatus [illegible] described [illegible] (198[illegible]). The test samples are thick-walled cylinders [illegible] mm in length with inner and outer diameters [illegible] and 12[illegible] mm, respectively. For ease of machining, each sample is in fact made up of three equally sized cylinders of rock. The rock samples are sealed in a leak-tight thin-walled copper jacket to exclude the pressure medium (argon). Temperatures are measured by eight thermocouples positioned at different radii in the sample. The large sample size and thermocouple redundancy (only two are necessary for a measurement) were chosen in order to increase the validity of the measurement on large-grained (>5 mm) rocks and to minimize the effects of local variation in a sample. The measurement is conducted inside an externally heated pressure vessel.

Heat flow is established in the sample in an approximately axisymmetric radial pattern by a small heater located along the axis of the sample. For the thermal conductivity measurement a steady-state heat flow is established. For the diffusivity measurement a transient heat flow condition, usually a 60-90 second pulse of the central heater, is imposed. Sample

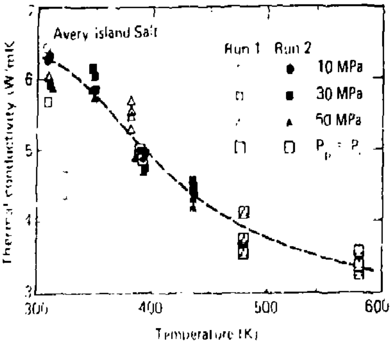

Figure 1. Thermal conductivity vs. temperature and pressure for two series of Avery Island salt. The error bar is typical of the random error (±1 s.d.) of each point. The dashed curve is hand drawn through the data.

length is monitored continuously. The equations used for data reduction are given by A[illegible] et al. (198[illegible]).

RESULTS

Rock Salt. Two samples (called runs 1 and 2 in the figures) of Avery Island rock salt were tested. The results for thermal conductivity, diffusivity, and thermal expansion are shown in Figs. 1-3, respectively. The first sample jacket failed near 473 K at 50 MPa, so [illegible] data at known effective pressure (defined as confining pressure minus pore pressure) are at room temperature (10 MPa to 50 MPa) and [illegible] K (50 MPa only). The jacket failure occurred because of the high thermal expansivity of the rock salt relative to the copper (4.5×10^{-5} K^{-1} vs. 1.6×10^{-5} K^{-1}). Once the jacket integrity was lost, the argon pressure medium was free to enter the pores and cracks of the sample and the effective pressure dropped below the confining pressure. The jacket leak was rather large and the measurements continued for several days, so it is assumed that the effective pressure fell to near zero. No change in the jacket design was made for the second rock salt sample, so in anticipation of another jacket failure between [illegible] and [illegible] K, temperature increments were made smaller and all pressures were explored (in the [illegible] MPa) at each new temperature. The second sample jacket failed at 438 K and 10 MPa. The first run was continued to its intended limit of 573 K following the jacket failure. The second run was terminated immediately following the jacket failure.

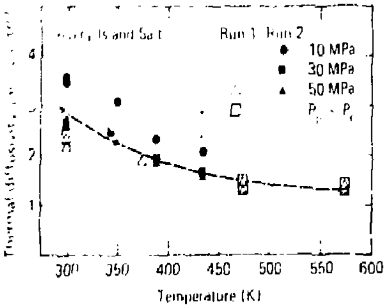

Figure 2. Thermal diffusivity vs. temperature and pressure for two samples of Avery Island salt. The same conventions apply as in Fig. 1.

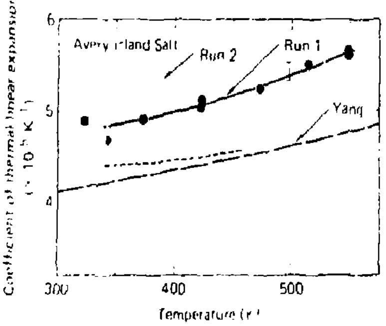

Figure 3. Thermal linear expansivity vs. temperature for two samples of Avery Island salt. Yang's curve is for single crystals and the short dashed curve represents our data if a system calibration is applied, that calibration being our measurements on quartz monzonite against the same measurements by Heard (1980).

The thermal conductivity (Fig. 1) and thermal diffusivity (Fig. 2) show good agreement between runs. Both decrease with increasing temperature. Thermal conductivity shows no discernable pressure dependence where the jacket is intact, and shows no obvious discontinuity when the jacket loses its integrity. Thermal diffusivity shows no dependence on pressure from 30 to 50 MPa, but apparently increases by approximately 20% when pressure drops to 10 MPa. The effect is seen in both runs. As discussed under ERROR the rise of diffusivity with decreasing pressure is tentatively explained away as a systematic error of unknown origin. The veracity of this explanation will be tested when a careful calibration of the apparatus is made.

The most extensive thermal expansion data (Fig. 3) came from the first run, where the temperature change was twice that of the second. The expansivity rises slightly with increasing temperature. The confining pressure for the first run was 50 MPa for all points, but above 473 K the pore pressure was also close to 50 MPa because of the jacket failure. The band of data in Fig. 3 from the second run represents repeated short (ΔT = 40 to 80 K) excursions upwards and downwards in temperature at several confining pressures. No dependence upon confining pressure was seen, nor was any temperature dependence seen, not surprising in light of the small temperature steps taken and the moderate temperature dependence detected in run 1. The width of the band compared to the height of the error bar for run 1 is an indication of the reproducibility of the thermal expansion data.

Quartz Monzonite. Two samples (runs 1 and 2) of Climax Stock quartz monzonite have been tested. Results for thermal conductivity, diffusivity, and thermal expansion are shown in Figs. 5-9. Thermal expansion data for the second run suggest apparatus problems and are not included. Testing plans for both samples called for repeated excursions to higher temperatures at the highest pressure (50 MPa) followed by heating under lower pressure. However, both runs were terminated prematurely during the 50 MPa heating cycle, both because of jacket failures. The first run produced conductivity data (Fig. 5) and diffusivity data to 150°C (Fig.8). The second run produced conductivity data to 250°C (Figs. 6,7) and diffusivity data to 200°C (Fig. 8). Absolute values do not agree well from run 1 to run 2 for reasons which may be the result of poor system calibration (see ERROR) rather than differences in the rocks themselves. However, the qualitative behavior of the thermal properties with changing P and T is consistent between runs.

Thermal conductivity (Figs. 5-7) shows a smooth decrease with increasing temperature and an accelerating decrease with decreasing pressure. The reduced scatter from run 1 to run 2 is the result of a change in the measurement technique (see ERROR). The improved precision of the measurement in run 2 has allowed resolution of a pressure effect (Fig. 7) and has revealed a curvature toward zero slope in conductivity vs. temperature. Neither run resolved any change in the room temperature conductivity vs. pressure as a result of heating under 50 MPa pressure, most clearly demonstrated in Fig. 7.

Thermal diffusivity is offset 50% from run 1 to run 2 for reasons which aren't yet known (see ERROR). Diffusivity in both runs decreases proportionally about the same amount with increasing temperature.

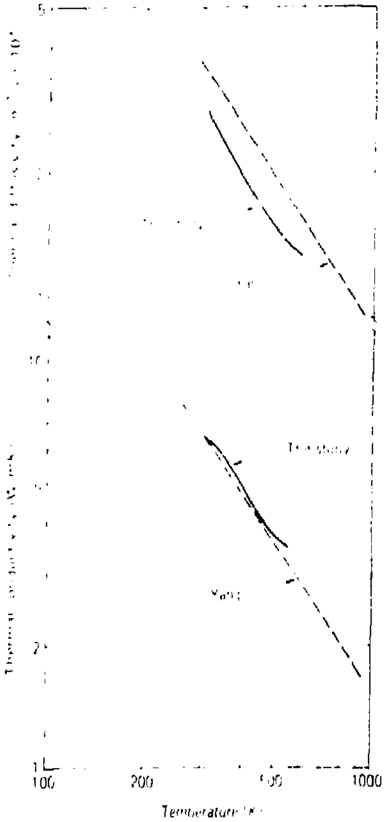

Figure 4. Thermal diffusivity and conductivity of Avery Island salt compared to Yang's single crystal values.

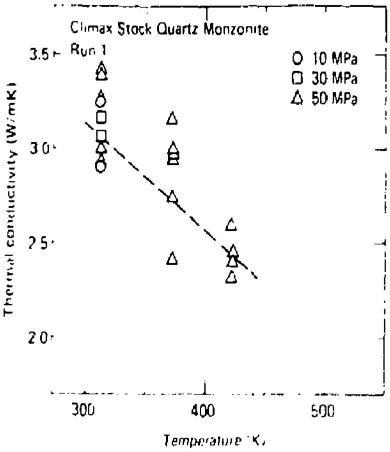

Figure 5. Thermal conductivity vs. temperature and pressure for the first run on Climax Stock quartz monzonite. The dashed curve is hand drawn.

The effect of thermal cycling was not checked, but was assumed to be the same as that for conductivity on the basis of the direct relationship between diffusivity and conductivity. The effect of pressure on diffusivity matches the anomalous behavior seen in salt: diffusivity is roughly unchanged from 30 to 50 MPa, but rises significantly at 10 MPa. As discussed below, the effect is suspected to be an artifact of the measurement system.

ERROR

All measurements reported here have been made without benefit of a system calibration, so their accuracy is somewhat speculative and best judged at this point by how they compare to existing values. Measurement resolution, discussed in the following paragraph, has only recently improved to the point where a useful calibration run, using a reference

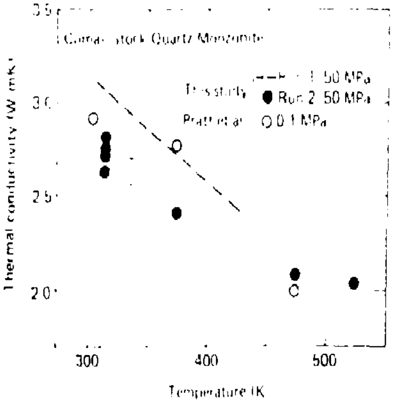

Figure 6. Thermal conductivity vs. temperature for two samples of Climax Stock quartz monzonite. The error bar is the typical random error of our run 2 data. The dahsed curve is lifted from Fig. 5.

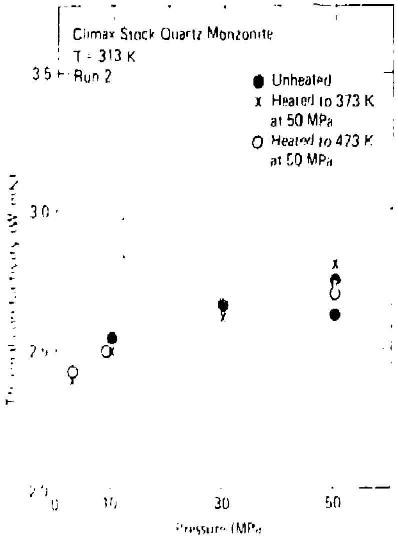

Figure 7. Thermal conductivity vs. pressure for Climax Stock quartz monzonite run 2 at 313 K following various heat treatments. The error bar ±3% is the estimated measurement precision.

standard, can be made.

Precision of the thermal conductivity measurement for salt runs 1 and 2 and for quartz monzonite run 1 is about ±0.3 W/mK and accounts for most of the scatter in Figs. 1 and 5. Most of this error is caused by ±0.1°C fluctuations in the outputs of three controlling thermocouples over the > 1 month duration of a run. A radical change in the measurement technique has improved precision for the quartz monzonite run 2 to about ±0.07 W/mK, estimated based on the scatter in Fig. 7 and on cursory parameter sensitivity studies made during run 2. The new measurement scheme replaces control of heater temperatures by control of heater powers, but requires computer modeling of the system in order to determine desired heater powers. One additional possible source of systematic error has therefore been introduced, so while the precision of the run 2 data is superior to that of the other data, their accuracy could be worse.

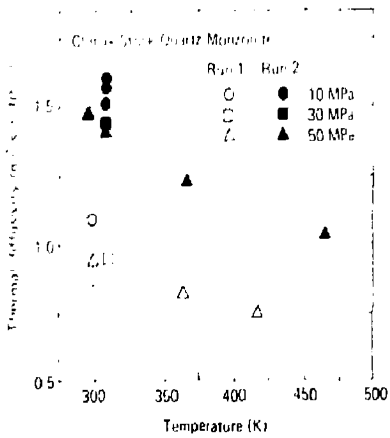

Figure 8. Thermal diffusivity vs. temperaure and pressure for two samples of Climax Stock quartz monzonite.

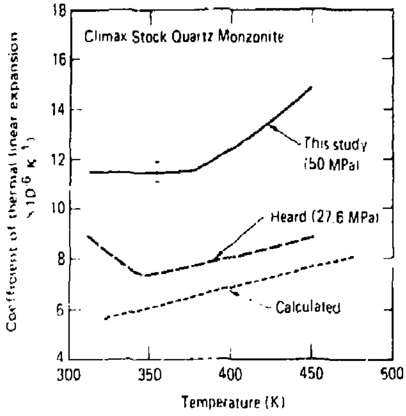

Figure 9. Thermal linear expansivity of Climax Stock quartz monzonite run 1. The calcualted curve is an estimate of the intrinsic rock expansivity based on a weighted average (by volume concentration) of the expansivities of the individual minerals in the rock.

The precision of the diffusivity measurement is no better than ±10% at 1.0×10^{-6} m^2/s. Whether the difference between Climax Stock quartz monzonite runs 1 and 2 (Fig. 8) is the result of poor precision or poor accuracy is uncertain. (It is probably not the result of sample differences because the thermal conductivity mismatch from run 1 to run 2 (Fig. 7) has a different sign than the diffusivity mismatch.) The anomalous rise in room temperature diffusivity at 10 MPa seen in all four runs is a sign that some systematic problems exist: given thermal conductivity vs. pressure (Figs. 1, 5, 7), the direct relationship between conductivity and diffusivity, and the fact that crack porosity tends to impede thermal transport, the expected change in diffusivity with pressure is either no change (if no cracks exist) or a decrease with decreasing pressure (if some cracks are present).

DISCUSSION

Rock Salt. For the salt we observe no pressure dependence of any of the thermal properties (excepting the anomalous behavior of diffusivity at 10 MPa) at about the ±5% resolution level (Figs. [illegible]). Based on the Walsh and Decker (1966) equation ([illegible]), a 10% change in thermal conductivity between 10 and 50 MPa (the maximum which would escape our detection at the ±5% resolution limit) corresponds to a change of rock volume due to cracks of roughly 0.13%. Even assuming all of this crack porosity was induced by heating to 573 K (i.e. assuming none of the original 1% porosity (Kaufmann, 1960) was crack porosity), it is a small value compared, say, to most granitic rocks. Heard (1980), for instance, sees in the Climax Stock quartz monzonite a thermally-induced porosity increase at 0.1 MPa of 0.4% between 300 and 573 K. Such a small amount of thermal damage in salt might be explained by its nearly isotropic expansivity (halite is cubic) and its low strength (Heard, 1972). The lack of pressure dependence of diffusivity (Fig. 2) and the smoothly increasing coefficient of thermal linear expansion (Fig. 3), uninterrupted by the jacket failure and drop in effective pressure, are also consistent with the existence of very little crack porosity.

The salt data are compared in Fig. 3 and Fig. 4 to an extensive compilation by Yang (1981) of single crystal NaCl data. The most serious disagreement is the diffusivity which falls about 20% below Yang's values at any given temperature. The apparent disagreement in thermal expansivity (Fig. 3) may vanish once system calibration is complete. If the correction required to bring the expansion data for the quartz monzonite into agreement with Heard's (1980) data (see Fig. 9) is applied to our rock salt data, the short dashed curve in Fig. 3 results. That curve is very close to Yang's.

Additional thermal conductivity measurements on rock salt for waste repository site-specific studies are reported by Morgan (1979) for two salts including Avery Island, and by Acton (1978) and Sweet and McCreight (1979) for the Southeastern New Mexico rock salt. These data are compared in Table 1. Only in the present study was true hydrostatic pressure applied to the test specimens. The data of Sweet and McCreight (1979) agree well with our data, Acton's (1978) data is scattered, and Morgan's Avery Island data is scattered but consistently much lower than our own. Although Morgan's samples were taken from a point in the mine horizontally separated from the source of our own by less than 50 m, he reports that the samples are weak and friable, rather in contrast to our own. A possible explanation of the different character of Morgan's samples is that his samples came from 50-mm diameter core while ours came from 400-mm core and that his samples were therefore more prone to damage during coring. It is plausible that the lower conductivities reported by Morgan are the result of excessive fractures and microfractures within his samples which were incurred during core drilling.

It should be pointed out that extensive testing of a similar rock salt, Southeastern New Mexico bedded salt, has demonstrated through mechanical testing (Wawersik and Hannum, 1980) and gas permeability measurements (Sutherland and Cave, 1980) that crack porosity in the starting material in the laboratory is non-zero. In both sets of experiments those authors conclude that the source of crack porosity is sample handling (coring, transport, experimental preparation) because the manifestations of crack porosity (non-linear pressure-volume relationship at lowest pressures, initial high permeability at low pressure) can be made to disappear permanently with the application of pressures in the 10-30 MPa range. Salt is known to deform plastically with relative ease at stresses of the same order (Heard 1972), so permanent crack closure under low pressures is plausible. In the present work, no measurements were made at zero effective pressure without first having subjected the sample to effective pressures of 10 MPa or more, so it is possible that any initial cracks which might have impeded heat transport in our samples were squeezed out of existence by the time the first measurements were made. The behavior of the Southeastern New Mexico salt and of the Avery Island salt appear to be consistent.

Quartz Monzonite. The quartz monzonite, in contrast to the salt, shows a pressure effect even in the unheated rock (Fig. 7), where the decrease in thermal conductivity is roughly 10% between 50 and 3 MPa. Taking the Walsh and Decker (1966) model, the required change in crack porosity is approximately 0.26%, a value which is consistent with a connected porosity (which presumably includes some round pores) of 0.54% (Page and Heard, 1981). Using the same analysis, the observation that heating to 473 K at 50 MPa produces no change, ±2%, in conductivity (Fig. 7) means that thermally induced crack porosity at 3 MPa, generated by heating to 473 K at 50 MPa, is less than 0.1%. The crack porosity increase under the same conditions as calculated by Page and Heard (1981) on the basis of linear expansion measurements is about 0.07%. It can at least be said that thermal conductivity vs. crack porosity as modeled by the Walsh and Decker equations (7) and (9), plus crack porosity vs. pressure as deduced by Page and Heard (1981) form a thermal conductivity vs. pressure relationship which, so far, is consistent with our own observations.

The only other data we are aware of for the conductivity of Climax Stock quartz monzonite at pressure and temperture are from Pratt et al. (1979). At room temperature they report a conductivity of 3.7 ± 0.2 W/mK with no measurable pressure dependence between 0.1 and 35 MPa. Although the magnitude is in serious disagreement with our measurements, our measured pressure-induced shift of 0.3 ± 0.1 W/mK beteen 3 and 30 MPa could be masked by the slightly lower resolution of the Pratt et al. measurement. Conductivity as a function of temperature at 0.1 MPa measured by Pratt et al. is plotted directly in our Fig. 6. The accelerating drop in conductivity with increasing temperature at 0.1 MPa which they observe is distinctly different from what we observe at 50 MPa. Presumably the rock at 0.1 MPa would be more prone to thermal cracking. The increase in crack porosity required by the Walsh and Decker model of the Pratt et al. data between 300 and 473 K is 0.8%, which does not agree well with the increase in crack porosity of 0.25% measured by Heard (1980) at 0.1 MPa.

The difference in thermal linear expansion (Fig. 9) between our data at 50 MPa and that of Heard (1980) at 27.6 MPa is probably more closely related to a lack of calibration of our system rather than the difference in pressure. Firstly, our 50 MPa curve indicates a higher expansivity than at 27.6 MPa, which is not reasonable. Recent data by Page and Heard (1981) show that at 55.2 MPa the expansivity of the quartz monzonite is lower than that at 27.6 MPa by 1-2 X 10^{-6} K^{-1} at all temperatures tested. Secondly, the expected intrinsic expansivity of the rock, calculated by weighing the expansivity of each phase by its volume concentration in the rock, falls close to Heard's curve at 27.6 MPa, suggesting that Heard's data are closer to the truth than our own. As mentioned above, if Heard's data is used to calibrate our thermal expansion measurement, our data on the thermal expansivity of salt also fall nicely into place with the intrinsic expansivity (Fig. 3).

Table 1

A comparison of laboratory measured thermal conductivities (in W/mK) for two rock salts.

	300 K	373 K	473 K	573 K
This study (AI)	6.3	5.3	4.0	3.3
Morgan (AI)	4.0	3.2	2.6	2.2
Acton (SENM)	8.5 3-8*	6.3		
Sweet & McCreight (SENM)	6.0	4.7	3.6	2.9

* for halite/anhydrite mixed-rock with > 50% halite. Conductivity increases with increasing purity of rock salt.

SUMMARY

Measurements of thermal conductivity, diffusivity, and linear expansion to 573 K and 50 MPa on two samples each of Avery Island salt and Climax Stock quartz monzonite have been carried out. From these measurements we conclude:

1) For Avery Island salt at $0 \leq P \leq 50$ MPa and $300 \leq T \leq 573$ K, the thermal conductivity, diffusivity, and linear expansion show no dependence (±5%) upon confining pressure. Thermal conductivity at room temperature measures 6.3 ± 0.6 W/mK and decreases monotonically to 3.3 ± 0.4 W/mK at 573 K. Thermal diffusivity decreases from $3.0 \pm 0.8 \times 10^{-6}$ m^2/s at room temperature to $1.4 \pm 0.5 \times 10^{-6}$ m^2/s at 573 K. Thermal linear expansivity increases from $4.8 \pm 0.3 \times 10^{-5}$ K^{-1} at room temperature to $5.6 \pm 0.3 \times 10^{-5}$ K^{-1} at 573 K. These values are approximately the intrinsic values for single crystal halite.

2) Handling of salt samples between the field and laboratory may damage the material sufficiently that zero pressure thermal conductivity of the bulk may be lowered by as much as 50%. The effect on conductivity can be permanently reversed by subjecting the rock to pressures of 10 MPa or more.

3) For Climax Stock quartz monzonite, room temperature thermal conductivity is approximately 3 W/mK with uncertain accuracy. Room temperature conductivity decreases at an accelerating rate by 0.28 ± 0.12 W/mK as pressure drops from 50 to 3 MPa. Within measurement error this behavior is not affected by heating at 50 MPa to temperatures up to 423 K.

4) In the quartz monzonite thermal conductivity drops approximately 0.7 ± 0.15 W/mK during 50 MPa isobaric heating from 300 to 523 K. Heating at lower pressures will probably cause a greater drop in conductivity due to thermally induced cracking.

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