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PREPRINT

THE THERMAL PROPERTIES OF ROCK SALT AND
QUARTZ MONZONITE TO 573 K AND 50-MPa
CONFINING PRESSURE

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to 573 K and 50-MPa Confining Pressure

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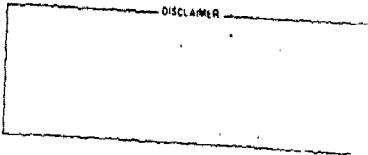
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Abstract

Measurements of thermal conductivity, thermal diffusivity, and thermal linear expansion have been made on two rock types, a rock salt and a quartz monzonite, at temperatures from 300 to 573 K and confining pressures from 10 to 50 MPa. The samples were taken from deep rock formations under consideration as possible sites for a nuclear waste repository--the rock salt from a domal salt formation at Avery Island, Louisiana, and the quartz monzonite from the Climax Stock, Nevada Test Site, Nevada. The testing temperature and pressures are meant to bracket conditions expected in the repository.

In both rock types, the thermal properties show a strong dependence upon temperature and a weak or non-dependence upon confining pressure. Thermal conductivity and diffusivity both decrease with increasing temperature in approximately linear fashion for samples which have not been previously heated. At 50 MPa in both rocks this decrease closely matches the measured or expected intrinsic (crack-free) behavior of the material. Preliminary indications from the quartz monzonite suggest that conductivity and diffusivity at low pressure and temperature may decrease as a result of heat treatment above 400 K.

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INTRODUCTION

The thermal properties of geological materials at elevated temperatures and pressures have attracted increased attention because of recent interest in disposing of nuclear waste in mined rock repositories. Accurate prediction of the long term response (over 10^4 - 10^5 years) of a waste repository to the thermal load of the waste requires the best possible determination of the physical properties of the repository rock at *in situ* physical and chemical conditions. In particular, knowledge of the thermal conductivity and diffusivity is required in order to model temperature distribution in time and space around the repository. The model in turn will help to predict expansions, stresses, microfracturing, and permeability changes in the repository.

Thermal properties data on rocks at room pressure abound, but because of experimental complexity and a lack of need, such data at pressures appropriate to the earth's upper crust are rather sparse. Hydrostatic pressure can be expected to have both an intrinsic and an extrinsic effect upon thermal conductivity and diffusivity in rocks: the intrinsic effect, because interatomic distances are changed, and the extrinsic effect, because of the presence of pores and microfractures. Some data have been taken in the intrinsic range, either at very high pressure where porosity has been squeezed out of existence or on single crystals where porosity doesn't exist. Bridgman (1) has shown that the intrinsic effect of pressure upon thermal conductivity is positive but quite small. In measurements to 1.2-GPa confining pressure he found the conductivity of NaCl increased linearly at a rate of 3.6% per 100 MPa, and that of basalt increased at a rate of 0.47%

per 100 MPa. The intrinsic effect of pressure on thermal diffusivity has also been measured on certain minerals by Kieffer et al. (2): for NaCl diffusivity increases at a rate of 3.1% per 100 MPa per $P \leq 1.8$ GPa; for quartz $\perp c$ diffusivity increases at a rate of 1.7% per 100 MPa for $P \leq 3.0$ GPa.

The effect of porosity on thermal conductivity has been modeled and confirmed by measurement to be substantial, with the magnitude of the effect dependent on the arrangement of the pore space (see Sweet (3) for a review). Walsh and Decker (4) related thermal conductivity to crack porosity theoretically and then used existing crack porosity vs. pressure relationships in order to construct a model of thermal conductivity vs. pressure. Experimentation (4) using uniaxial stress to approximate hydrostatic pressures supported the model well: intrinsic and bulk thermal conductivity differed by as much as 15% in dry rock at zero pressure. The effect of pressure parallels the pressure dependence of crack porosity. That dependence is qualitatively similar for most dense, crystalline rocks: strongest at zero pressure, decreasing steadily with increasing pressure to the intrinsic dependence where, presumably, all cracks have closed. The extrinsic effect becomes insignificant, depending on rock type, at pressures of 50 to 300 MPa (see, for instance, Brace (5)). The location of a waste repository, somewhere between the surface (lithostatic $P = 0$) and 1 to 2 km deep ($P = 25$ to 50 MPa), puts the repository rock where the extrinsic pressure effect is expected to be very strong. Therefore, the application of room pressure thermal properties measurements to the repository situation must be considered suspect.

If such measurements are made at elevated temperature, the application to a repository becomes suspect for yet another reason. It is well known that heating a rock, uniformly or otherwise, can lead to microfracturing (Richter and Simmonds (6); Cooper and Simmonds (7); Johnson et al. (8); Johnson and Gangi (9)). The cause is thermal expansion mismatches in a polymineralic matrix, whose mineral components themselves are (in most cases) anisotropic in their expansivity. The application of pressure would close some

of these cracks or, if the heating were done under pressure, might even suppress some of the microfracturing altogether.

The present work reports the first results from a newly built apparatus designed to measure thermal conductivity, diffusivity, and linear expansion under simultaneous elevated temperature and hydrostatic (gas) confining pressure. Results are reported for two rocks, a rock salt and a quartz monzonite.

SAMPLE MATERIAL

The rocks studied here are site specific. The rock salt samples were taken from a mine at Avery Island, Louisiana in one of several rock salt formations under consideration for a waste repository. The quartz monzonite was cored at a location adjacent to the site of a test spent fuel repository in the Climax Stock at the Nevada Test Site, Nevada.

The rock salt samples were cut from a 400-mm diameter core (C-11), supplied by RE/SPBC Inc., Rapid City, South Dakota. The core was taken from the floor of workings at the 163-m level at the site of an *in situ* heater test in the Avery Island domal salt formation. Chemical analyses of samples from the Avery Island Mine show roughly 99.1% NaCl (by weight), 0.7% water insolubles, 0.2% CaSO_4 (anhydrite), and 0.2% water (Kaufmann [10]). 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 [11]). 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 quartz monzonite has been characterized in detail by Izett (12) and Maldonado (13). Average composition of several samples taken from the U-15-A drill hole roughly 300 m horizontally distant from the spent fuel repository was determined as 28% by weight quartz, 25% alkali feldspar, 40% plagioclase, 6% biotite, and 1% accessory minerals (13). Grain size in the matrix is 1-1 1/2 mm but is marked by quartz phenocrysts (5-10% by volume) averaging 4 mm in diameter and by large orthoclase phenocrysts (5% by volume) averaging 50 mm in length with some as long as 150 mm. The orthoclase phenocrysts are uniformly distributed in the rock. A laboratory-sized sample measuring 50 cm^3 or more in volume which does not encounter an orthoclase phenocryst is unusual. The rock has a porosity of approximately 1% and has a density of approximately 2.64 Mg/m^3 .

EXPERIMENTAL TECHNIQUE

The measurement apparatus is described by Abey et al. (14). The samples to be measured are thick-walled cylinders 203 mm in length with inner and outer diameters of 19 mm and 127 mm, respectively. For ease of machining, each sample is made up of three equally sized cylinders of rock. The samples are clad in a leak-tight, thin-walled copper jacket to exclude the pressure medium (argon) from the rock. Temperature is measured by eight thermocouples positioned at eight 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 of large-grained ($>5 \text{ mm}$) samples 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- to 90-s pulse of the central heater, is imposed. Sample length is monitored continuously.

Because of the likelihood that thermal expansion, changes in permeability, and changes in thermal properties share at least one common mechanism (microfracturing), there is a distinct need to understand as much as possible about how reversible changes (such as opening and closing of existing microfractures) and irreversible changes (such as creation of new microfractures) affect the thermal properties. For this reason, measurements are made at pressures (P) and temperatures (T) bracketing those expected in a repository. The path in (P, T) space that the experiment follows is chosen so that microfracturing is postponed as much as is reasonable. Thus, (low P , low T) and (high P , low T) are explored before (high P , high T) and all are explored before (low P , high T).

RESULTS

Rock Salt

Two samples of Avery Island rock salt were tested. The results for thermal conductivity, diffusivity, and linear expansion are shown in Figs. 1-3, respectively. The first sample burst its jacket near 473 K and 50 MPa, so the only data at known effective pressure (defined as confining pressure minus pore pressure) are at room temperature (10 to 50 MPa) and 373 K (50 MPa only). The jacket burst occurred because of the high thermal expansivity of the rock salt relative to the copper ($4.5 \times 10^{-5} \text{ K}^{-1}$ vs. $1.0 \times 10^{-5} \text{ K}^{-1}$). Presumably, once the jacket integrity was lost, the pressure medium was free to enter the pores and cracks of the sample and the effective pressure dropped well below the confining pressure. The jacket leak was rather large, 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; thus, in anticipation of another jacket failure between 373 and 473 K, temperature increments were made smaller and all pressures were explored (in the order 50, 30, 10 MPa) at each new temperature. The second sample jacket failed at 438 K and 10 MPa. The first run was

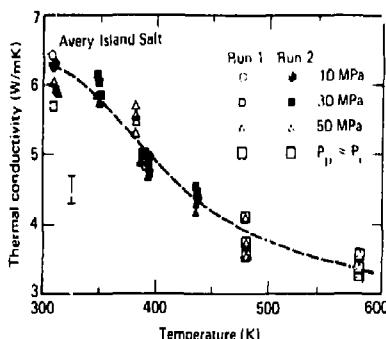


Fig. 1. Thermal conductivity vs. temperature and pressure as measured on two samples of Avery Island rock salt. The large squares around the higher temperature points of Run 1 indicate the measurements were made following jacket failure (see text). The error bar indicates the typical range of random error (one standard deviation). The $\text{d} - \text{d}$ curve is drawn free-hand through the data.

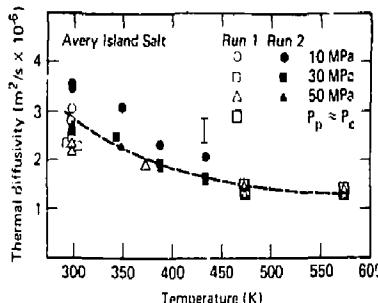


Fig. 2. Thermal diffusivity vs. temperature and pressure as measured on two samples of Avery Island rock salt. The same conventions apply as in Fig. 1.

continued to its intended limit of 573 K following the jacket failure. The second run was terminated immediately following the jacket failure.

The thermal conductivity (Fig. 1) and thermal diffusivity (Fig. 2) show good agreement between runs, both decrease strongly with increasing temperature. Thermal conductivity shows no discernible 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 and at all temperatures tested. Based on the direct relationship between conductivity and diffusivity, the same pressure effect (i.e., none) would be expected for each. Based on the microcrack model either no pressure dependence (rock has no microcracks) or a decrease of diffusivity with pressure (rock has microcracks) would be anticipated. The rise of diffusivity with decreasing pressure is therefore considered anomalous and is tentatively explained away as a systematic error. 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

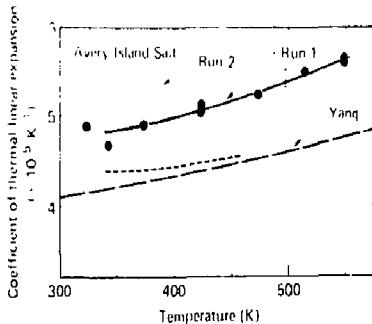


Fig. 3. Thermal linear expansivity of two samples of Avery Island salt. Yang's (22) curve is the expansivity of single crystal halite. The short dashed curve represents our data if a system calibration is applied, that calibration being our measurements on quartz monzonite against Heard's (18) measurements on the same rock (Fig. 6).

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, so the data indicate no dependence of expansivity upon effective pressure between 300 and 573 K. The band of data in Fig. 3 from the second run represents repeated short ($\Delta T = 40$ to 80 K) excursions upward and downward 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

One sample of Climax Stock quartz monzonite has been tested to date, and a jacket failure (caused this time by a reaction between the copper jacket and pyrite, FeS_2 , inclusions in the rock) terminated that run at the same point, and after the same traverse in (P, T) space as the first rock salt run. The run was terminated immediately following the jacket failure. The results for thermal conductivity, diffusivity, and linear expansion are given in Figs. 4-6, respectively.

The thermal conductivity of the quartz monzonite (Fig. 4) at 50 MPa shows a distinct decrease with increasing temperature. Values at any given temperature are approximately half those for rock salt. At room temperature there is no discernible dependence of thermal conductivity upon pressure for $10 \text{ MPa} \leq P \leq 50 \text{ MPa}$. The room temperature pressure independence remained even following the excursion to 423 K at 50 MPa. Thermal diffusivity (Fig. 5) also decreases with increasing temperature at 50 MPa, shows no pressure dependence at room temperature for $30 \text{ MPa} \leq P \leq 50 \text{ MPa}$, and shows the same anomalous rise at 10 MPa as rock salt, reinforcing the notion that the rise represents system problems.

The coefficient of thermal linear expansion during a single heating episode from room temperature to 468 K is shown in Fig. 6. The rate of expansion is independent of temperature from 320 K to approximately 375 K and increases steadily with temperature from 375 to 468 K.

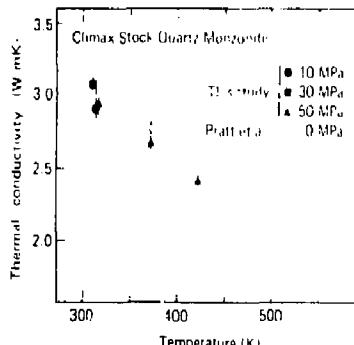


Fig. 4. Thermal conductivity of Climax Stock quartz monzonite vs. temperature and pressure.

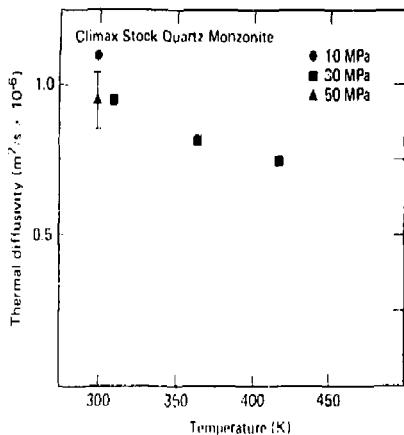


Fig. 5. Thermal diffusivity of Climax Stock quartz monzonite as a function of temperature and pressure.

DISCUSSION

The thermal properties of rock salt as measured here (ignoring the anomalous behavior of diffusivity at 10 MPa) are not dependent upon confining pressure. In light of the suggested role of microcracks in rocks, such behavior might be expected only in a monomineralic rock (such as rock salt) whose single phase is isotropic. Halite has cubic symmetry and may be close enough to isotropic that significant (from the point of view of heat storage and transport) thermal cracking does not occur. The data gathered here are strongly self-consistent (again excepting diffusivity at 10 MPa), given the well-understood and demonstrated relationship between microfracturing and enhanced thermal

expansivity (Richter and Simmons (6); Cooper and Simmons (15); Bruner (16); Wong and Brace (17); Heard (18)). The smoothly increasing coefficient of thermal linear expansion, uninterrupted by the jacket rupture and drop in effective pressure, indicates that uniform heating of rock salt generates very little motivating force for microfracturing. The thermal conductivity in such a material could only be expected to show no extrinsic pressure dependence.

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 (19)) and gas permeability measurements (Sutherland and Cave (20)) that crack porosity in the starting material in the laboratory (as opposed to *in situ*) is non-zero. In both sets of experiments, those authors conclude that the source of crack porosity is sample handling (core, transport, experimental preparation) because the manifestations of crack porosity (nonlinear 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 differential stresses of the same order (see, for example, Carter and Heard (21)), so permanent crack closure under low pressures is plausible. In the present work, no measurements were made at zero effective pressure without the sample first being subjected to effective pressures of 10 MPa or more. Therefore, 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 rock salt and of the Avery Island rock salt appear to be consistent.

The salt data are compared in Fig. 3 and Fig. 7 to an extensive compilation by Yang (22) 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 (18) data (see Fig. 6) 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 (23) for two salts including Avery Island, and by Acton (24) and Sweet and McCreight (25) 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 (25) agree well with our data, Acton's data (24) are scattered, and Morgan's Avery Island data are 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 is that his samples came from 50-mm-diameter core while ours came from 400-mm core. 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.

The thermal properties data on quartz monzonite are too incomplete for detailed analysis at this point. Thermal conductivity shows no pressure dependence at room temperature, even following heating to 423 K at 50 MPa. This behavior would suggest that the

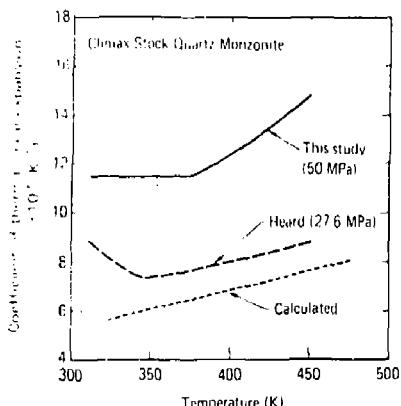


Fig. 6. Thermal linear expansivity of Climax Stock quartz monzonite at 50 MPa. Data are compared to Heard's (18) measurements at 27.6 MPa on the same rock type and to an "intrinsic" expansivity calculated by averaging the expansivities of the individual minerals making up the rock, weighted according to their volume percentage in the rock.

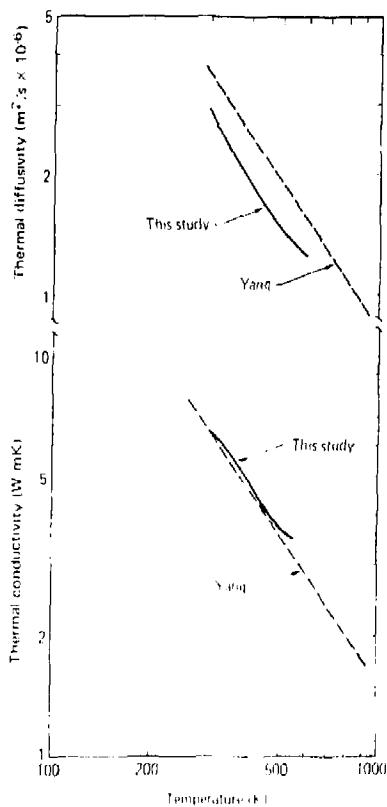


Fig. 7. Thermal conductivity and diffusivity of Avery Island rock salt measured here compared to single crystal data compiled by Yang (22).

1% porosity reported for the Climax Stock quartz monzonite (13) does not apply to our sample, or that the porosity is tied up in round pores which are not strongly influenced by pressures below 50 MPa, or that the porosity is tied up in very flat cracks which close completely at pressures below 10 MPa. Pressure-volume relationships measured for the granodiorite in the Climax Stock, a close sister to the quartz monzonite, show no evidence of such a high (i.e., 1%) crack porosity influenced by pressure below 10 MPa (26). In fact, the hydrostat shown by Schock et al. (26) contrasted with that of the Casco granite used by Walsh and Becker (4) in an application of their model is revealing: the Casco granite shows a crack porosity of 0.4% by the Walsh and Becker definition (i.e., where a straight line extension of the intrinsic portion of the P-V curve intersects the volume strain axis); the P-V curve for the Climax Stock granodiorite, using the same definition, shows a crack porosity of well under 0.05%. The 0.4% crack porosity in the Walsh and Becker model leads to a predicted 15% change in thermal conductivity for dry rock between 0 and 100 MPa. If all other factors in the Walsh and Becker equation (7) are unchanged, a 0.05% crack porosity would mean only a 2% change in conductivity for the quartz monzonite.

TABLE 1. Comparison of thermal conductivity measurements.

	Thermal conductivity (W/mK)			
	300 K	373 K	473 K	573 K
This study (AI)	6.3	5.3	4.0	3.3
Morgan (AI) (23)	4.0	3.2	2.6	2.2
Acton (SENM) (24)	8.5 3-8 ^a	6.3	---	---
Sweet & McCreight (SENM) (25)	6.0	4.7	3.6	2.9

^aFor halite/anhydrite mixed-rock with >50% halite (conductivity increases with increasing purity of rock salt).

One other set of thermal conductivity measurements is available for the quartz monzonite (27) and these are shown in Fig. 4. The Pratt et al. (27) data were taken at 0-MPa effective pressure, and where there is overlap (room temperature and 373 K) the agreement with our data is good. A higher temperature point at 473 by Pratt et al. falls distinctly below the trend of the lower temperature data and may reflect microfracturing.

Microfracturing is anticipated in the quartz monzonite at higher temperatures and lower pressures on the basis of theoretical considerations mentioned above and on the basis of the thermal expansion results of Heard (18). Heard's curve for 27.6-MPa pressure is shown in Fig. 6 for comparison with our own data at 50 MPa. At lower pressures, Heard finds systematically higher expansivities and suggests that these are caused by microfracturing.

The difference in thermal linear expansion (Fig. 6) between our data at 50 MPa and that of Heard (18) at 27.6 MPa is probably more closely related to a lack of calibration of our system rather than to the difference in pressure. First, our 50-MPa curve indicates a higher expansivity than the 27.6-MPa, which is not reasonable. Second, 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 are 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).

SUMMARY AND CONCLUSIONS

From thermal properties measurements carried out at elevated effective confining pressure and elevated temperature on Avery Island rock salt and Climax Stock quartz monzonite, both of interest for the country's nuclear waste isolation effort, we conclude:

1) For $0 \leq P \leq 50$ MPa and $300 \leq T \leq 573$ K, the thermal conductivity, diffusivity, and linear expansion of the rock salt 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) At room temperature, neither unheated quartz monzonite nor quartz monzonite heated as high as 423 K at 50 MPa shows any effect of confining pressure upon thermal transport behavior. The thermal expansivity of the rock at 50 MPa for $100 \leq T \leq 437$ K is approximately its intrinsic expansivity.

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