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THERMAL CONDUCTIVITY AND DIFFUSIVITY OF CLIMAX STOCK QUARTZ MONZONITE AT HIGH PRESSURE AND TEMPERATURE

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ABSTRACT

Measurements of thermal conductivity and thermal diffusivity have been made on two samples of Climax Stock quartz monzonite at pressures between 3 and 50 MPa and temperatures between 300 and 523 K. Following those measurements, the apparatus was calibrated with respect to the thermal conductivity measurement using a reference standard of fused silica. Corrected thermal conductivity of the rock indicates a value at room temperature of 2.60 ± 0.25 W/mK at 3 MPa increasing linearly to 2.75 ± 0.25 W/mK at 50 MPa. These values are unchanged (± 0.07 W/mK) by heating under 50-MPa pressure to as high as 473 K. The conductivity under 50-MPa confining pressure falls smoothly from 2.75 ± 0.25 W/mK at 313 K to 2.15 ± 0.25 W/mK at 473 K. Thermal diffusivity at 300 K was found to be $1.2 \pm 0.4 \times 10^{-6}$ m^2/s and shows approximately the same pressure and temperature dependencies as the thermal conductivity.

INTRODUCTION

The desire to measure the thermal properties of rocks at upper crustal conditions of pressure (<100 MPa) and temperature (<500°C) has been stimulated by recent interest in permanent storage of radioactive waste materials in mined underground repositories. An apparatus has been designed and built to make such measurements on large-grained (to approximately 10 mm) polycrystalline rocks (Abey et al., 1981). Earlier papers reported measurements made by this apparatus on rock salt (Durham et al., 1981) and preliminary results on a quartz monzonite from the Climax Stock, Nevada Test Site (Durham and Abey, 1981). Those papers also discussed in detail the rationale for making measurements at in situ conditions of pressure and temperature and the expected thermal response of crystalline rock to changes in pressure and temperature.

This report presents the latest measurements of thermal conductivity and diffusivity for Climax Stock quartz monzonite (CSQM) and presents a recent calibration of the test apparatus. The CSQM data presented here are the first calibrated data we have reported. For a detailed description of the apparatus and experimental method, as the expected thermal response of a model polycrystalline solid containing microfractures, the reader is referred to the earlier papers.

TEST SAMPLES

Two samples of CSQM (referred to hereafter as Run 1 and Run 2) were tested and a calibration test was made using a sample of fused silica with known thermal conductivity. The mineralogy of the CSQM is detailed by Izett (1960). Briefly, the average composition of core taken from the U-15-A drill hole approximately 300 m horizontally distant from the site of the spent fuel test is 28% by weight quartz, 25% alkali feldspar, 40% plagioclase, 6% biotite, and 1% accessory minerals. Grain size in the matrix is 1 to 1.5 mm, but is marked by quartz phenocrysts (5 to 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 volume of rock 50 cm^3 or more that does not encounter an orthoclase phenocryst is unusual. The rock has a connected porosity of approximately 0.54% (Page and Heard, 1981) and has a density of approximately 2.64 Mg/m^3.

The fused silica reference standard was taken from a single ingot of General Electric Type 124 clear fused silica. Details of the chemistry and of the physical properties of the material are available on product data sheets from the manufacturer. It is a high-purity silica glass (approximately 54 ppm impurity atoms, 50% of those being Al) with a low coefficient of thermal linear expansion (0.55×10^{-6} K^{-1}). The low expansivity makes it an ideal pressure calibration standard, given the apparent absence of materials in the conductivity range 1 to 10 W/mK whose pressure dependencies over the range 0 to 50 MPa are well-established. Its resistance to thermal shock makes it unlikely to develop fractures (or microfractures) with a concurrent extrinsic (crack-related) dependence of any of its physical properties upon pressure. The intrinsic pressure dependence in silica glass is negligible over the range 0 to 50 MPa (Bridgman, 1952, for conductivity, 0.38%/100 MPa; Kieffer et al., 1976, for diffusivity, -0.10%/100 MPa), so it is assumed for the purposes of calibration that the dependencies of the several thermal properties upon temperature remain unchanged in the pressure range 0 to 50 MPa. The temperature dependence of thermal conductivity at atmospheric pressure is based on independent measurements made on a piece of fused silica cut from the original ingot, immediately adjacent to the center section of our reference standard (Fig. 1).

MEASUREMENT AND CORRECTION PROCEDURES

For all runs, measurement of thermal diffusivity and, for CSQM Run 1, measurement of thermal conductivity were made by the procedure given in Abey et al., 1981. The measurements of thermal conductivity for CSQM Run 2 and for the fused silica reference standard were made by a significantly different technique, which is outlined below. Because the two techniques are different, the calibration run was used to correct only data taken for CSQM Run 2.

The main point of difference between the two techniques is the control of heater powers rather than the control of heater temperatures. The heaters referred to are the three inner sample heaters used to maintain the temperature gradients that allow conductivity to be measured (Fig. 1). The temperatures are those of the controlling thermocouples, also shown in Fig. 1. Of the multitude of heat

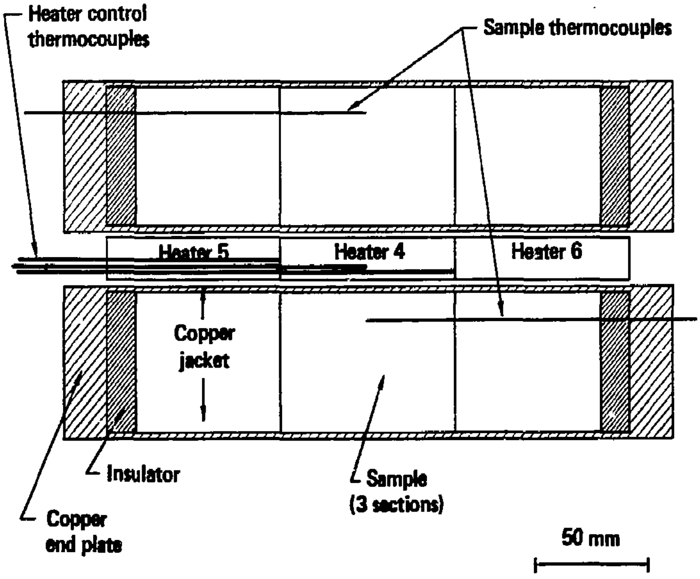

Fig. 1. Sample assembly cross section.

flow patterns that can be generated in the sample with three independently controllable heaters, any one may be selected by either of the two methods: heater powers may be set to desired levels (how the desired levels are decided upon is discussed below), or heater powers may be set so that thermocouple readings lie at desired levels. In the ideal situation of perfect instrument precision, no meaningful distinction can be made between the two methods. It is a characteristic of the experimental configuration, however, that measured conductivity is considerably more sensitive to thermocouple temperature fluctuation than heater power fluctuation. At 3 W/mK, a temperature fluctuation of ± 0.1 K at 300 K shifts measured conductivity by approximately ± 0.3 W/mK. To produce the same shift, heaters operating at, say, 3 W must fluctuate approximately 0.2 W. Given standard measurement equipment, therefore, heater control is a more practical means of achieving satisfactory measurement precision of thermal conductivity.

The desired heater power levels are those that produce an apparent conductivity (a function of the radial temperature gradient and output power from the central of the three heaters, as given in Abey et al., 1981) equal to the actual conductivity. These levels are determined by a numerical simulation of the heat flow in the measurement apparatus. The desired power condition is invariably one in which the two end heaters operate at the same power, which in turn is somewhat higher than the power output of the central heater (Fig. 1). The ratio of the output of one end heater to that of the central heater is hereafter referred to as the power ratio. Good accuracy of the numerical model is assured by calibrating it against a sample of known thermal conductivity. Obviously, the closer the conductivity of the known standard to that of the unknown rock, the more accurate will be the calibrated model as applied to the unknown rock. In the present case, the difference between the conductivity of the CSQM (near 3 W/mK at 300 K) and that of the fused silica (near 1 W/mK at 300 K) is significant. The possible effect on the accuracy of the final conductivity measurement is discussed below.

In actual fact, the CSQM Run 2 was made prior to the run on the reference standard, and previously published data from CSQM Run 2 (Durham and Abey, 1981) were reduced using an uncalibrated model. Correction after the fact is made possible by determining the behavior of the model and the real system with changes in the power ratio (defined above). As shown below, tests on the reference standard indicate remarkable agreement between the model and real system as regards the change of apparent conductivity with power ratio. Correcting the prior measurement of the unknown, therefore, is simply a matter of using the calibrated model to determine what should have been the apparent conductivity had the improper power ratio been applied (which was the case). The conductivity of the model is then adjusted until the apparent conductivity of the calibrated model matches the apparent conductivity during the run. At that point, the modeled conductivity becomes the best estimate of the real conductivity of the rock. In future situations, with the calibrated model in hand, the proper ratio will be known and will be applied during the measurement.

RESULTS

Uncorrected Data

Figure 2 shows uncorrected values of thermal conductivity as a function of temperature for both CSQM samples. The reduced scatter between Runs 1 and 2 is the result of the change in measurement procedure discussed in the previous section. Figure 3 gives uncorrected thermal conductivity as a function of pressure at 313 K (Run 2 data only) and Fig. 4 shows uncorrected thermal diffusivity as a function of temperature and pressure for both runs.

Calibration of the Model

Figure 5a illustrates the main features of the calibration procedure. Apparent conductivities were measured on the fused silica calibration standard at room temperature at each of four confining pressures (3, 10, 30, 50 MPa) at several values of power ratio between 1.7 and 2.9. The results are the points with error bars in Fig. 5a. The (uncalibrated) model which was used to predict the power ratios for CSQM Run 2 was applied to the calibration standard, with the results enclosed in the small oval in Fig. 5a. The

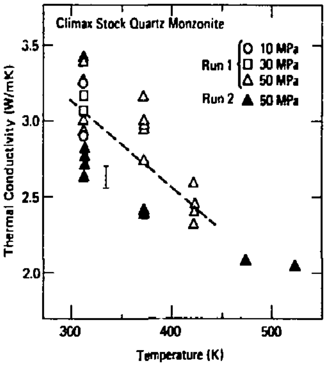

Fig. 2. Thermal conductivity vs. temperature for CSQM Runs 1 and 2, uncorrected. Dashed line is drawn freehand through the data of Run 1 and is transposed to Fig. 6.

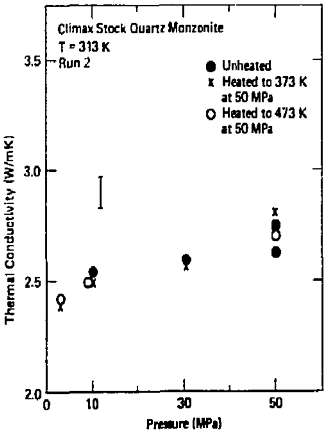

Fig. 3. Thermal conductivity vs. pressure at 313 K for CSQM Run 2, uncorrected.

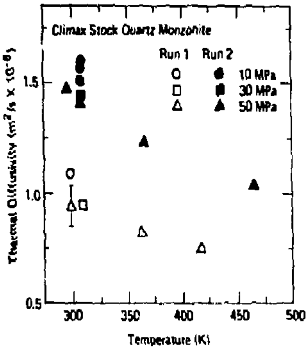

Fig. 4. Thermal diffusivity vs. temperature and pressure for CSQM Runs 1 and 2, uncorrected.

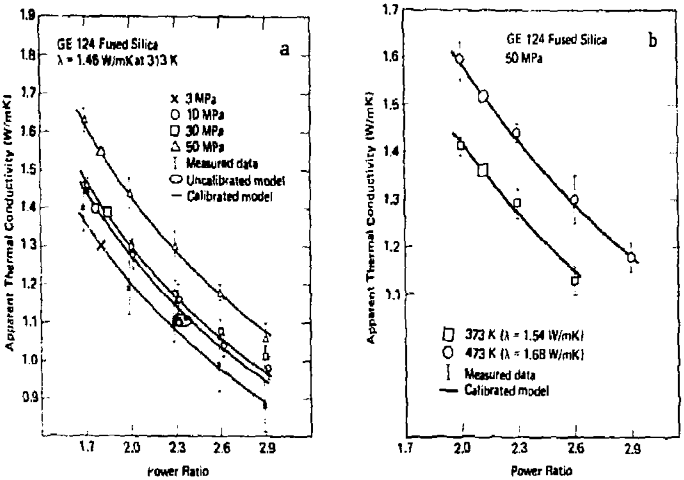

Fig. 5. Thermal conductivity calibration data for fused silica reference standard a) for various pressures, and b) for various temperatures (see text).

predicted power ratios are nearly constant, between 2.32 and 2.36, for all confining pressures. The data in Fig. 5a show that application of those power ratios results in conductivities that vary considerably with pressure (as can be seen by projecting vertically from the small oval to the actual data). If, in fact, the thermal conductivity of the fused quartz is constant at 1.46 W/mK as we are assuming, then the uncalibrated model must be inaccurate.

Figure 5b shows the data for the calibration standard at elevated temperature and fixed pressure. At the time the data were taken, the true conductivity was not known, but was assumed to be in the range 1.2 to 1.4 W/mK. Therefore, none of the measured (apparent) values of conductivity in Fig. 5b reached the true value.

Every attempt was made in the construction of the original model to faithfully copy the physical details of the apparatus. The observation of relatively subtle differences between expected and actual behavior in the fused silica (Fig. 5a) is an indication of the success of the original model. In calibrating the model, two parameters were varied: a) the conductance across the inner jacket-rock interface, infinite in the uncalibrated model, was allowed to decrease with decreasing confining pressure; b) the convective heat transfer from the outer metallic surfaces of the sample assembly, originally perfect, was allowed to decrease with decreasing confining pressure. The adjustment of these two parameters was done manually until the model matched, within an arbitrary error, the observed behavior of the fused silica. The calibrated model in its final form produces the solid lines shown in Figs. 5a and b.

Corrected Data

Following the procedure described above under "Measurement and Correction Procedures," the CSQM Run 2 conductivity values in Figs. 2 and 3 were corrected as shown in Figs. 6 and 7, respectively. Calibration of the thermal diffusivity measurements has not yet been accomplished.

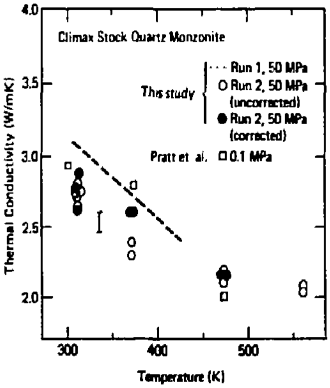

Fig. 6 Thermal conductivity vs. temperature at 50 MPa for CSQM Run 2, corrected and uncorrected. Dashed line is uncorrected data from Run 1, taken from Fig. 2. Error bar indicates precision of data from this study.

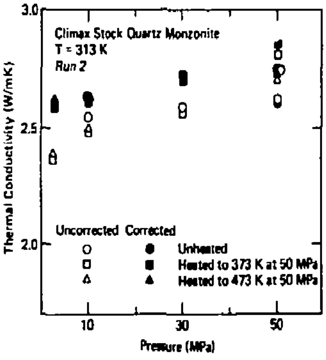

Fig. 7. Thermal conductivity vs. pressure at 313 K for CSQM Run 2, corrected (solid symbols). Also shown is the uncorrected data from Fig. 3 (open symbols).

DISCUSSION

The data in Fig. 7 indicate an approximately linear increase in thermal conductivity of CSQM with increasing pressure between 3 and 50 MPa. The best estimate of conductivity at 313 K is 2.60 ± 0.25 W/mK at 3 MPa and 2.75 ± 0.25 W/mK at 50 MPa. The error bands include a measurement precision (reproducibility) of ± 0.07 W/mK (± 1 std. dev.) and an estimated uncertainty in the accuracy of the model of ± 0.20 W/mK. Within the measurement precision, the pressure dependence at 313 K is unchanged by heating under 50-MPa confining pressure to temperatures as high as 473 K, as shown by Fig. 7. The variability in thermal conductivity from sample to sample cannot be determined from our data. Independent measurements of the thermal conductivity of CSQM at 300 K by Pratt et al. (1979) indicate a value of 3.7 W/mK without mention of accuracy. Within a measurement precision of ± 0.2 W/mK (D. Enniss, 1980), Pratt et al. found the thermal conductivity at 300 K to be unchanged by the application of confining pressure over the range 0.1 to 35 MPa.

Under 50-MPa pressure, thermal conductivity changes smoothly with temperature in approximately 1/T fashion (Fig. 6), the usual intrinsic behavior of materials in the phonon conduction range (Touloukian and Ho, 1981). Data at 0.1 MPa from Pratt et al. (1979), also plotted in Fig. 6, show a sharper drop in conductivity with increasing temperature than our own and are difficult to explain by intrinsic conduction mechanisms alone. One possibility is that the Pratt et al. samples, without the benefit of confining pressure, have developed microfractures upon heating which in turn have acted to inhibit heat transport.

Thermal diffusivity of CSQM (Fig. 4) shows a similar dependence upon confining pressure (little or none) and temperature (approximately 1/T) to that of thermal conductivity, again the expected behavior for a material whose thermal capacity (density times heat capacity) does not change significantly over the range of pressure and temperature encountered here. Most rocks in fact have a thermal capacity within 20% of 2.3 X 10^{-6} Ws/m^3K (Touloukian and Ho, 1981). That value, applied to the CSQM data in Fig. 6 and using the relationship

$$\text{Thermal diffusivity} = \frac{\text{thermal conductivity}}{\text{thermal capacity}}$$

produces values intermediate to those of the two runs in Fig. 4. No explanation can be given for the strong disparity between the diffusivity results for Runs 1 and 2 except to say that the precision of the diffusivity measurement has not yet been refined as well as the precision of the conductivity measurement.

CONCLUSIONS

1. The thermal conductivity of as-cored CSQM at 313 K varies in approximately linear fashion with increasing pressure from 2.60 ± 0.25 W/mK at 3 MPa to 2.75 ± 0.25 W/mK at 50 MPa. These values are not altered more than ± 0.07 W/mk by heating under 50-MPa confining pressure to as high as 473 K. These values are based on measurements of a single 127-mm-diameter by 203-mm-long volume of rock. The stated error band includes absolute uncertainty.

2. Under 50-MPa confining pressure, thermal conductivity of CSQM varies approximately as 1/T, the expected intrinsic behavior.

3. The thermal diffusivity of CSQM at 300 K and 50 MPa is 1.25 ± 0.4 X 10^{-6} m^2/s. Within a precision of ± 0.1 X 10^{-6} m^2/s, the variation of diffusivity with pressure and temperature is proportional to that of thermal conductivity. The proportionality factor is the density times the heat capacity.

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