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The Thermal Conductivity of Talc as a Function of Pressure and Temperature[1]

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Talc is a commonly used pressure-transmitting and gasket material for high-temperature and -pressure applications. The thermal conductivity of talc at high pressures and temperatures is therefore valuable in the design of high-pressure experiments and apparatus. In this paper measurements of the thermal conductivity of fired and unfired talc are presented. Measurements were made at pressures ranging from 0 to 2.5 GPa and temperatures from 150 to 900 K. The thermal conductivity was measured with the hotwire technique. The thermal conductivity results for both the fired and the unfired talc show a slight increase with increasing pressure. The absolute value of the thermal conductivity of talc is lower in the fired material than in the unfired material. In both cases, the thermal conductivity varied less than 15% over the temperature range studied. X-Ray diffraction studies have shown talc to be highly disordered. The results are shown to be consistent with those expected for a disordered crystal.

KEY WORDS: high pressure; hot-wire method; talc; thermal conductivity.

1. INTRODUCTION

Talc is a commonly used pressure-transmitting and gasket material at high temperatures and pressures. It is readily available, easily machined, and a good thermal and electrical insulator. In addition, its mechanical properties make it particularly suited to these applications [1, 2].

In high-temperature, high-pressure work, a sample is invariably surrounded by a solid pressure transmitter and gasket-forming materials. If temperatures greater than a few hundred degrees are required, the heat is normally generated internally by means of electrical heating. It is thus important to know the thermal conductivity of the materials used to form

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gaskets and to transmit the pressure to the sample since the sample holder must protect the walls of the pressure vessel from the intense heat generated inside it. To do this, the thermal resistance of the materials used must be significantly higher than that of the walls of the pressure vessel.

The thermal conductivity of talc and its variation with pressure and temperature are therefore useful parameters in the design of high-temperature, high-pressure experiments and apparatus. Although detailed studies of the mechanical properties of talc have been made, the thermal properties have been neglected in comparison. The availability of a high-temperature, high-pressure hot-wire technique in our laboratory capable of covering the range 0–2.5 GPa and 150–900 K makes it possible to measure the thermal properties under conditions closely approximating those used in practice.

The thermal conductivity of talc was measured as a function of pressure and temperature. Dehydrated materials are frequently used in high-temperature applications to prevent sample contamination from the water released at high temperatures. Therefore, in addition to the measurements made on the unfired material, measurements were made on talc fired to 1173 K for 2 h. Talc loses 80% of its chemically bound water at 1173 K [3]. The results presented for the fired material are therefore strictly valid for use only when experimental temperatures are below 1173 K. Talc becomes very hard when fired above 1173 K and hence loses its effectiveness as a pressure transmitter.

2. EXPERIMENTAL

The thermal conductivity was measured using the hot-wire technique described previously [4–6]. This is a dynamic technique in which a current pulse is applied to a metal wire embedded in the sample material. The wire itself is used as the temperature probe. The temperature rise of the wire as a function of time is related to the thermal properties of the surrounding material by the equation:

$$\lambda = \frac{q d \ln T}{4\pi\, dt}$$

where q is the power per unit length in the hot-wire, T is the temperature, t is the time, and λ is the thermal conductivity.

The talc used in these experiments was obtained in powder form from Ferro Industrial Products, South Africa. The compositional specifications are given in Table I. The unfired talc samples were prepared directly from the powder as received, by pressing into tablets 18 mm in diameter and 5 mm long at a pressure of 0.2 GPa. The unfired talc was white in color.

Table I. Specifications of Talc Type EW45

SiO_2	61.73
MgO	30.43
CaO	0.38
Al_2O_3	0.85
Fe_2O_3	0.89
K_2O	0.02
Na_2O	0.06

Loss on ignition, 5.3

The fired talc samples were prepared by firing the loose powder at 1173 K for 2 h. The resultant powder was beige in color. Samples were then prepared in the same way as for the unfired material. The fired material was difficult to compact and tended to flake when it was removed from the die. In both the fired and the unfired talc samples the fine powder fraction $<100\ \mu m$ was used. X-Ray analysis has shown that this technique results in pellets with no preferred orientation. The hot wire was sandwiched between two cylinders of the sample material. It was arranged in a semicircle on the sample surface to ensure that the pressure distribution was uniform over its length.

Two materials were used for the hot wire. Below room temperature, 99.95% pure Ni wire, 0.127 mm in diameter was used. Nickel, however, has a Curie point at 633.1 K. This results in a large change in the temperature coefficient of resistance in the range of the Curie point and makes thermal conductivity measurement impossible over a range of about 100 K. Platinum is suitable for high-temperature measurements but pure Pt wires failed to survive pressure cycling. To resolve this problem, a composite hot wire was constructed with a Pt wire and Ni leads as shown in Fig. 1. The

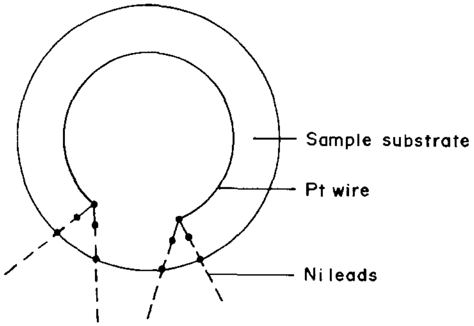

Fig. 1. Diagram of composite hot wire on sample surface.

only possible complication resulting from the use of a composite wire is the effect of thermal emf's generated at the junction of two dissimilar metals. However, by reversing the current direction in two consecutive measurements, this effect was found to be negligible provided that the sample temperature was stable. The resistance was measured by a four-point method and therefore the presence of Ni in the leads did not affect the resistance measured for the Pt wire.

A schematic diagram of the sample capsule used is shown in Fig. 2. The capsule is constructed of fired pyrophyllite, which is a solid pressure-transmitting medium and ensures that the sample experiences a pressure which is close to hydrostatic. The pyrophyllite is fired prior to use to avoid sample contamination by the water which would otherwise be released at high temperatures. The sample temperature was raised by passing a current through the internal graphite heater and could be varied from room temperature to 900 K. The graphite heater shown in Fig. 2 has a stepped configuration to minimize the temperature gradient along the sample. Finite-element analysis was used to optimize the heater design. The gradient was less than 3 K up to temperatures of 900 K for the configuration used. The copper cylinder at the top of the cell reduced the heat dissipated in this

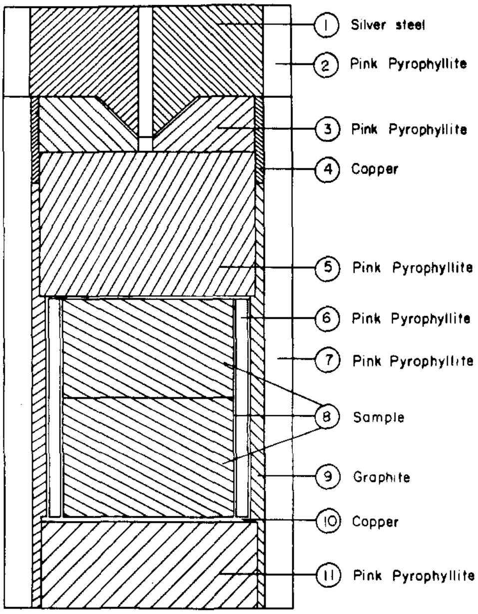

Fig. 2. Schematic diagram of high-temperature, high-pressure capsule.

region and prevented excessive extrusion of the components at the top of the cell and the resultant breakage.

The sample temperature was lowered by cooling the whole pressure vessel with a controlled flow of liquid nitrogen. The sample temperature was deduced from the change of resistance of the metal wire, after correcting for wire deformation as described by Andersson and Bäckström [6]. The temperature coefficient of resistivity was obtained in a separate experiment at high pressure for high-purity nickel and platinum. The temperature coefficient was found to be independent of pressure for both the Ni and the Pt wires used.

A piston-cylinder apparatus was used to generate the pressure. The cell was located in the bore of a 30-mm pressure vessel and pressure was applied by the rams of a 30 KN Kennedy press. The pressure was measured by a load cell located below the piston. Friction corrections were made to the nominal pressure based on the hysteresis measured using a manganin resistance gauge. The pressure calibration has been found to be accurate to within 0.05 GPa [7].

3. RESULTS

3.1. Pressure Dependence

The thermal conductivity of fired and unfired talc was measured as a function of pressure at ambient temperature between 0 and 2.5 GPa. The samples were precompacted at 2 GPa before the commencement of the measurements to ensure good contact between the sample and the hot wire.

The results are shown in Fig. 3 for fired and unfired material respectively. Regression fits were made to the experimental data and the results are given in Table II. In both cases the thermal conductivity increases with increasing pressure. The absolue value of the thermal conductivity is significantly lower in the fired material than in the unfired material.

The only data available in the literature for comparison are those of Hughes and Sawin [8]. They report measurements on talc after heating to 1273 K for 3 h to remove the waters of hydration. They found a rapid increase in thermal conductivity from 0.7 $W \cdot m^{-1} \cdot K^{-1}$ at zero pressure and 488 K to 13 $W \cdot m^{-1} \cdot K^{-1}$ at 0.2 GPa and 444 K. These results are an order of magnitude higher than those obtained for fired talc in the present experiment. Closer examination of the results of Hughes and Sawin for KCl obtained with the same technique shows that their results are very different from those of Andersson [9]. It therefore seems likely that a systematic error was distorting their results.

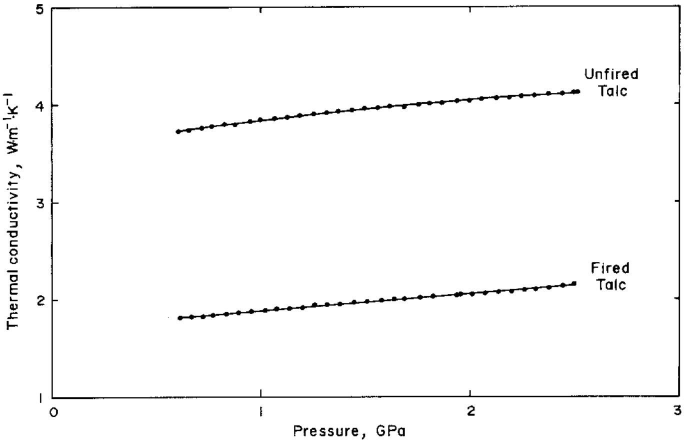

Fig. 3. Variation of the thermal conductivity of fired and unfired talc as a function of pressure at ambient temperature. The solid lines are regression fits to the experimental data.

The pressure dependence of the thermal conductivity is generally discussed in terms of the parameter g, where

$$g = \left(\frac{\partial \ln \lambda}{\partial \ln \rho}\right)_T = B_T \left(\frac{\partial \ln \lambda}{\partial P}\right)_T$$

where λ is the thermal conductivity, ρ the density, P the pressure, and B_T the isothermal bulk modulus of talc [10]. The bulk modulus of unfired talc may be determined from the compressibility data of Vaidya and Kennedy [11] and is given by

$$B_T = 36.95 + 0.309\ P + 0.0388\ P^2$$

where B_T is the isothermal bulk modulus and P is the pressure in GPa.

The g value obtained from the present experiment for unfired talc is 2.62. Unfortunately, no data exist for the bulk modulus of fired talc and therefore no value for g can be calculated in this case.

Table II. Pressure Dependence of Thermal Conductivity of Fired and Unfired Talc: Coefficients of Thermal Conductivity Fitted to Equations of the Form $\lambda = a(0) + a(1) + a(2)\ P^2$

Material	$a(0)$ (W m^{-1} K^{-1})	$a(1)$ (W m^{-1} K^{-1} GPa^{-1})	$a(2)$ (W m^{-1} K^{-1} GPa^{-2})
Unfired talc	3.540	3.369×10^{-1}	-4.290×10^{-2}
Fired talc	1.701	1.747×10^{-1}	—

3.2. Temperature Dependence

The temperature dependence of thermal conductivity was measured at 2.5 GPa in the range 300 to 900 K. In each case the temperature dependence was recorded at the highest pressure that could be reached in the apparatus (2.5 GPa) so that the measuring conditions were as close as possible to those under which talc is typically used in high-pressure experiments. The results are shown in Fig. 4.

The results for unfired talc show a linear decrease with increasing temperature. To confirm this trend, a second run was performed on the unfired material in the temperature range 200–300 K. The results agreed with those recorded at high temperatutres as can be seen in Fig. 4. It has been observed that the structure of talc remains unchanged up to temperatures of 973 K during which time only adsorbed water is lost. Above this temperature, interlayer water is lost and the material structure is modified. The results for the unfired material may therefore be extrapolated only to 973 K [3].

The results for fired talc show a linear increase with increasing temperature in the range 300–900 K (Fig. 4). These results may be extrapolated to 1173 K but further phase changes [3] above this temperature prevent further extrapolation.

Linear regression fits were made to the experimental data and the results are given in Table III for the unfired and fired material, respectively.

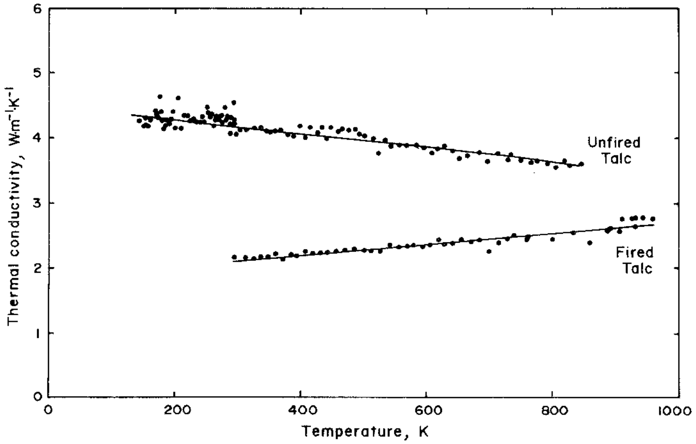

Fig. 4. Variation of the thermal conductivity of fired and unfired talc as a function of temperature at 2.5 GPa. The solid lines are regression fits to the experimental data.

Table III. Temperature Dependence of the Thermal Conductivity of Fired and Unfired Talc at 2.5 GPa: Coefficients of the Thermal Conductivity Fitted to Equations of the Form $\lambda = b(0) + b(1)\,T$

Material	$b(0)$ ($W\,m^{-1}\,K^{-1}$)	$b(1)$ ($W\,m^{-1}\,K^{-2}$)
Unfired talc	1.849	8.455×10^{-4}
Fired talc	4.498	-1.096×10^{-2}

4. DISCUSSION

Unfired talc has a double-layer monoclinic (pseudohexagona) structure with a brucite layer located between two hexagonal silica layers; the structural formula is $Mg_3(Si_2O_5)_2(OH)_2$ [12]. Lindemann [13, 14] has shown that the dehydroxylation of talc at 1223 K leads to the formation of protoenstatite and amorphous silica. Upon cooling, the protoenstatite transforms to enstatite, and the silica to critobalite.

The bulk of theoretical effort in the field of thermal conductivity has been devoted, on the one hand, to materials with simple crystal structures, e.g., cubic crystals, and on the other hand, to completely amorphous materials. The complex crystal structure of talc therefore falls beyond the scope of this theory and detailed discussion of the results is difficult. However, some interesting parallels may be drawn between the results observed and the theoretical predictions.

A structurally perfect, harmonic crystal would have zero thermal resistivity. In an anharmonic crystal, thermal resistivity arises from phonon–phonon interactions. A given phonon can interact with other phonons whose number density at high temperatures is proportional to temperature, so the thermal conductivity is inversely proportional to the temperature [15]. This predicted temperature dependence is strictly valid only for isochoric conditions but has been found to exist for isobaric conditions when the effect of thermal expansion is relatively small. The general trend for the pressure dependence of structurally perfect crystals is an increase in thermal conductivity with increasing pressure.

The above discussion applies only to perfect crystalline materials. Ross et al [16] have proposed that in the case of a small amount of structural disorder, we may assume that the thermal resistivity caused by the disorder simply adds to that due to phonon–phonon interactions. Hence

$$W = A(\mathrm{st}) + A(\mathrm{ph})\,T$$

The quantity $A(\mathrm{st})$ is a temperature-independent term associated with structural disorder and $A(\mathrm{ph})$ is a coefficient of the Liebfried–Schlömann type. For the pressure dependence both terms need to be considered, but in practice $A(\mathrm{st})$ seems to be relatively insensitive to pressure. This explanation is consistent with the small absolute value and weak temperature dependence found experimentally for disordered crystals, e.g., plastic crystal phases [16]. Typical g values for disordered crystals are of the order of 4.

It is well known from X-ray diffraction that talc commonly exhibits very disordered stacking (see, e.g., Ref. 17). The study by Akizuki and Zussman [18] confirms that the structure is disordered on the unit-cell scale. The small temperature dependence of the thermal conductivity observed for unfired talc may therefore be explained if one considers the structural disorder term to dominate the Liebfried–Schlömann term in the above expression. The low g value observed is also consistent with this explanation.

For glasses and amorphous materials, the thermal conductivity is found to increase slightly with increasing temperature. Similar behavior has been found for some crystalline but disordered materials, e.g., clathrate hydrates [16]. Daw et al. [12] report from transmission electron microscopy that the dehydroxylation of talc occurs inhomogeneously, with dislocations acting as preferential sites for the dehydroxylation. The resulting enstatite crystallites are randomly oriented. The disorder in the fired talc is therefore greater than that in the unfired talc due to a partial transformation of the structure. It is therefore possible that the increasing thermal conductivity with temperature observed in the case of the fired talc may be due to an increased degree of disorder in the fired sample. The g values of glasses and amorphous materials are typically in the range 1–4. The small pressure dependence observed for the fired talc as well as the small absolute value of thermal conductivity supports the description of the fired material as a highly disordered system.

5. CONCLUSIONS

The thermal conductivity of fired and unfired talc compacted powder samples has been measured as a function of pressure and temperature in the range 0–2.5 GPa and 150–900 K. The thermal conductivity was found to be fairly insensitive to both pressure and temperature in the range studied. However, the thermal conductivity of the unfired material decreased with increasing temperature, while that for the fired material increased with increasing temperature. This behavior can be attributed to a large extent to the high degree of disorder of the structures of these materials.

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