Thermal conductivity of ME771 glass-epoxy laminate from millikelvin temperatures to 4 K

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Permaglas ME771 is a glass-epoxy laminate which is suitable for use at cryogenic temperatures. We have measured the thermal conductivity of a sample of this material between 64 mK and 4.2 K in the direction parallel to the reinforcing fibres, enabling us to make a comparison with the better known material G-10CR. The thermal conductivity follows the form that would be expected for such a material, and is similar to that of G-10CR, which has a similar (room temperature) tensile strength. We comment on some confusion that has arisen over the difference between G-10CR, a material specifically produced for cryogenic use, and G-10, the more common equivalent.

Keywords: Structural materials (A), Thermal conductivity (C), Instrumentation (D)

1 Introduction

Polymers are commonly used at cryogenic temperatures when structural components are required with low thermal conductivity. The strength of polymers can be considerably increased by mixing them with fibres which provide reinforcement. There are many different forms of fibre-reinforced polymer (FRP) commercially available, differing in the materials used both for the polymer and the fibre components. These materials generally offer a low κ/E ratio [1] (where k is the thermal conductivity and E is Young's modulus) and low thermal expansivity [2], making them suitable for rigid supports with good thermal isolation [3–6]. However, as with most classes of material, the properties of FRPs at cryogenic temperatures are known only for a handful of materials. This lack of knowledge is greatest at temperatures below 1 K — a temperature range at which knowledge of material properties is particularly important since cooling power is generally extremely limited.

In order to extend such knowledge, we have measured the thermal conductivity of Permaglas ME771, a red coloured oriented glass epoxy laminate produced by Röchling[1]. The operating temperature range is given as -270 °C to 155 °C. As with most such materials, the reinforcing fibres are laid in planes, and the properties of the material depend on whether they are measured parallel or perpendicular to this plane. The greatest strength is obtained parallel to the fibre direction. We made our measurements in this direction; the datasheet tensile strength at room temperature is 450 MPa.

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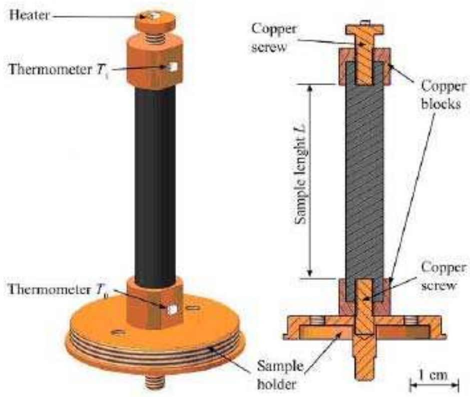

Figure 1: Set up for the measurements of the samples. A second thermometer (not shown here) is mounted at each end of the sample, as described in the text.

2 Experimental technique and measurements

The thermal conductivity was measured by the longitudinal steady heat flow method, using our standard technique [7]. A known power P was supplied to one end of the sample to establish a difference of temperature $T_1 - T_0$ along the sample. By differentiation of the integrated power (at constant T_0)

$$P(T_1) = \frac{A}{L}\int_{T_0}^{T_1} \kappa(T)dT \tag{1}$$

the thermal conductivity $k(T)$ was obtained, where A and L are respectively the sample section and length.

The experimental set up for the thermal conductivity measurement is shown in Fig. 1. Thermal contact was made to each end of the sample both by tightly fitting copper cups around the ends and 4 mm diameter copper screws threaded into the samples, ensuring good thermal contact no matter what the relative thermal contraction of copper and the sample is. The copper blocks at the two ends of the sample each had a RuO_2 thermometer (used at temperatures below 2 K) and a Cernox thermometer (for temperatures above 2 K) glued onto them. An SMD (surface mount device) NiCr heater was mounted on the "warm" end of the sample.

The electrical connections to the heater and to the thermometer were made with 25 µm diameter NbTi wires, electrically connected by tiny crimped Cu tubes. At the ends of the NbTi wires a four lead connection was adopted. The bottom copper block was screwed onto a copper sample holder in thermal contact with the mixing chamber of a dilution refrigerator. The thermometers were calibrated by means of a SRD 1000 (Superconductive Reference Device) and an NBS - SRM 767a fixed point device [8–10]. The sample was enclosed by a copper shield in thermal contact with the mixing chamber of the dilution refrigerator. The thermometers were measured using an AVS 47 a.c. resistance bridge and power to the heater was supplied by a four wire I - V source meter (Keithley 2601).

The sample was a cylinder with diameter of (8.00 ± 0.02) mm. The length (over which the temperature gradient was measured) was $L = (89.68 \pm 0.05)$ mm giving a room temperature geometrical ratio of $g = A/L = (0.561 \pm 0.006)$ mm. Our measurements of conductance include the thermal impedance between the copper blocks and the sample. To ensure that this was negligible, we carried out a second measurement on a sample with about twice the geometrical ratio of the initial sample. The same values for conductivity were obtained within experimental error.

2.1 Details of calculating $k(T)$

Thermal conductivity was obtained by differentiation of $P(T_1)/g$, where P is the applied power and T_1 is the temperature of the warm end of the sample.

There are three main contributions to the relative error in $k(T)$:

– the power supplied to the sample: we estimate that the relative error of P is of the order of $\sim 0.1\%$;
– the measurement of the form factor g. The error in the measurements of g is estimated to be less than 1%;
– the uncertainty in the temperature, dT, due to the accuracy of the thermometers in this temperature range. A conservative value of $(dT)/T$ is $\sim 2\%$ for $T > 1K$ and $\sim 1\%$ for $T < 1K$.

Taking into account these contributions, the maximum relative error in $k(T)$ is about 3%.

3 Results and discussion

The measured thermal conductivity is shown in Fig. 2 and Table I. The conductivity values can also be expressed (in $Wm^{-1}K^{-1}$) as:
$k(T) = (12.0 \pm 0.1)10^{-3}T^{(2.10\pm0.01)}$ for $T < 600$ mK
$k(T) = (8.76 \pm 0.06)10^{-3}T^{(1.524\pm0.009)}$ for $600\ \mathrm{mK} < T < 4.2$ K

In the measured temperature range, we expect the overall conductivity to depend on the contact resistance between the fibres and the epoxy as well as the conductivity of the two components [11]. As the details of the composition of ME771 are proprietary and unknown to us we are unable to attempt to predict the thermal conductivity in a quantitative way, but we can compare the measurements with other similar materials.

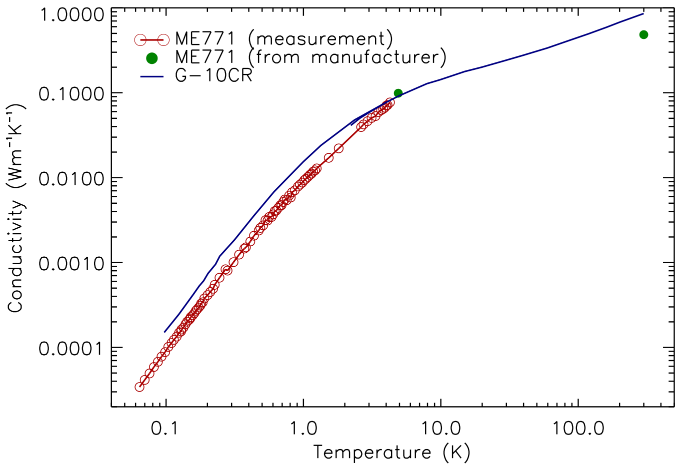

Figure 2: The measured thermal conductivity of ME771 (○). Also shown: data from the manufacturer (•), and values for G-10CR (solid line); these consist of two independent measurements in overlapping temperature ranges [16, 17], with good agreement seen between them. All measurements are made parallel to the fibre direction.

Temperature (K)	Conductivity ($Wm^{-1}K^{-1}$)
0.064	3.42×10^{-5}
0.070	4.15×10^{-5}
0.080	5.62×10^{-5}
0.100	9.18×10^{-5}
0.125	0.000149
0.150	0.000219
0.200	0.000412
0.300	0.000911
0.400	0.00166
0.500	0.00264
0.600	0.00365
0.700	0.00485
0.800	0.00585
1.00	0.00908
1.25	0.0129
1.50	0.0167
2.00	0.0257
2.50	0.0361
3.00	0.0479
3.50	0.0587
4.00	0.0701
4.26	0.0768

Table I: Measured conductivity of ME771 (the measured values are interpolated to a set of temperatures at fixed intervals).

Probably the most popular fibre reinforced polymer used at cryogenic temperatures is a glass fibre reinforced epoxy commonly known as G-10. This is generally used for printed circuit boards, and is usually green in colour. A "cryogenic grade", called G-10CR, is also available. There appears to be considerable confusion over the meaning of the terms G-10 and G-10CR, and we take the opportunity here to attempt to explain the situation. It is somewhat misleading to refer to G-10CR as the cryogenic grade of G-10, since G-10 is not the name of a material; it is a NEMA[2] specification describing electrical and mechanical properties (this standard is not freely available, but the military equivalent [12] is). Considerable latitude is left as to the actual composition, with the specification stipulating only that it should consist of "glass cloth or nonwoven parallel aligned fibres" in an "epoxy-resin compound or binder". Different materials are sold as meeting the G-10 specification, and are commonly referred to as G-10 (or FR-4, a fire retardant replacement, which is itself often incorrectly called G-10/FR-4). The specification does not specify Young's modulus or tensile strength, and says nothing about thermal conductivity, even at room temperature. It is therefore likely that samples of G-10 from different manufacturers will have different properties in these areas.

To avoid this problem, the G-10CR specification was developed by NIST.[3] The specification describes particular source materials and processing [15] to produce a well defined material which meets the G-10 specifications, made with an epoxy which has proven performance at cryogenic temperatures. Not only has the thermal conductivity been measured over the temperature range from 0.1 K to room temperature, values for the tensile strength and Young's modulus are also known [14, 15]. The likelihood of variability in G-10 means that measurements which have shown that particular samples of G-10 have similar properties to G-10CR do not mean that everything sold as G-10 can be considered to be a (cheaper and more readily available) equivalent to G-10CR. Indeed, there is some evidence of G-10 with considerably higher thermal conductivity than expected for G-10CR [13].

We therefore compare our measurements on ME771 with G-10CR, not G-10; this is shown in Fig. 2. In this Figure, we supplement our measurements on ME771 with thermal conductivity values (also parallel to the fibre direction) at around 5 K and room temperature supplied by the manufacturer. The 5 K value is in good agreement with our results. It can be seen that the conductivity of ME771 is similar to G-10CR. The tensile strength at room temperature is also similar; the datasheet value for ME771 (parallel to the fibre direction) is 450 MPa, while values of 431 ± 27 MPa are reported for C-10CR [14]. We compare room temperature values since this is all that is available for ME771; the tensile strength of G-10CR increases with decreasing temperature [15], and this is presumably also the case for ME771. The manufacturer's datasheet for ME771 does not specify Young's modulus, so we cannot compare this with G-10CR. While other materials (particularly carbon fibre epoxies) are available with better strength to conductivity ratios in this temperature range [18], ME771 would appear to be a useful alternative to G-10 and G-10CR, with the advantage over G-10 that it is a properly specified material.

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