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THERMAL CONDUCTIVITY MEASUREMENTS OF EPOXY SYSTEMS AT LOW TEMPERATURE.

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ABSTRACT

We have developed a specific thermal conductivity measurement facility for solid materials at low temperature (LHe and LN2). At present, the Measurement of Thermal Conductivity of Insulators (MECTI) facility performs measurements on epoxy resin, as well as on bulk materials such as aluminium alloy and on insulators developed at Saclay. Thermal conductivity measurements on pre-impregnated fiber-glass epoxy composite are presented in the temperature range of 4.2 K to 14 K for different thicknesses in order to extract the thermal boundary resistance. We also present results obtained on four different bonding glues (Stycast 2850 FT, Poxycomet F, DP190, Eccobond 285) in the temperature range of 4.2 K to 10K.

INTRODUCTION

Epoxy resin and fiberglass composites are used in the manufacturing of superconducting magnets of large dimensions, in particular for the electrical insulation and the mechanical bonding of conductors in the winding of well-known “fully epoxy-impregnated coils”. The anisotropic constitution and the various elaboration modes of these resin-fiberglass end in a large disparity in their physical properties, in particular in their thermal properties at low temperatures. For the large magnet projects where inadequate design lead dramatic consequences in terms of performances, cost and time, each “non-traditional” material used at low temperatures must be carefully tested. For this reason, our laboratory, involved in the construction of several magnets for LHC, has developed a thermal conductivity experiment (MECTI) to characterize different materials as pre-impregnated fiberglass-epoxy composite and epoxy glues around liquid helium temperature.

Nowadays, commercial epoxy glues are used in the construction of superconducting coils, as thermal linkage and structural bonding for the cooling circuits. For some of theim, the thermal conductivities have been measured [1,2]. Others products could be potentially interesting, however their thermal properties are not systematically given in the range of 4 to 15 K. To examine their possibilities, we have retained 4 glues to be tested with MECTI.

PRINCIPLE OF THE MEASUREMENT

Our thermal conductivity measurements are based on a steady-state method with a longitudinal heat flow. Heat flow is then considered as one-dimensional along the sample. Pure conduction Fourier's law is given by equation (1) :

$$Q = \frac{S}{l} \cdot \int_{Tc}^{Th} \lambda(T).dT \qquad (1)$$

where Q is the heat flux, S the transversal section (assumed constant), l the sample length, T_c and T_h the cold and the hot temperature of the sample, respectively, and $\lambda(T)$ its thermal conductivity. Average thermal conductivity λ_{av} between T_c and T_h is

$$\lambda_{\mathrm{av}} = \frac{1}{(T_h - T_c)} \cdot \int_{Tc}^{Th} \lambda(T).dT = \frac{Q \cdot l}{S \cdot (T_h - T_c)} \qquad (2)$$

If λ is a linear function of T, equation (2) is simplified to :

$$\lambda_{\mathrm{av}} = \lambda[\frac{T_h + T_c}{2}] = \frac{Q \cdot l}{S \cdot (T_h - T_c)} \qquad (3)$$

As experimental results will show, the thermal conductivity of our resin is not a linear function of T. Nevertheless, for small temperature differences and small variations of λ (T) (as measured), the equation (3) can be applied in our measurements and the induced error is less than 1 to 2 % [3].

DESCRIPTION OF THE SET-UP

As already carried out in other laboratories, our apparatus is developed to measure thermal conductivity in steady-state conditions[4]. The sample is located between a cold heat sink cooled by a cryogenic bath (here, liquid helium used) on one side and a heat source on the other as described in Figure 1. Heat flow is assumed to be transverse and one-dimensional between the two faces of the sample. High vacuum around the sample stops heat leak via convection and the use of small diameter thermalized manganin wires for instrumentation further limits thermal losses by conduction. Conduction losses are also minimized on the sample holder by using nylon and G10 pieces behind the heater and stainless steel tie rods. Radiation heat transfer is negligible around liquid helium temperature because vacuum tank is immersed in the liquid He bath. Thermal differential dilatations are compensated by using Belleville washes. Heat flow losses from heater outside sample are less than 0.3 % in the 0-55 mW range . Accuracy on difference temperature measurement is about +/- 8 mK in the 4-15 K range.

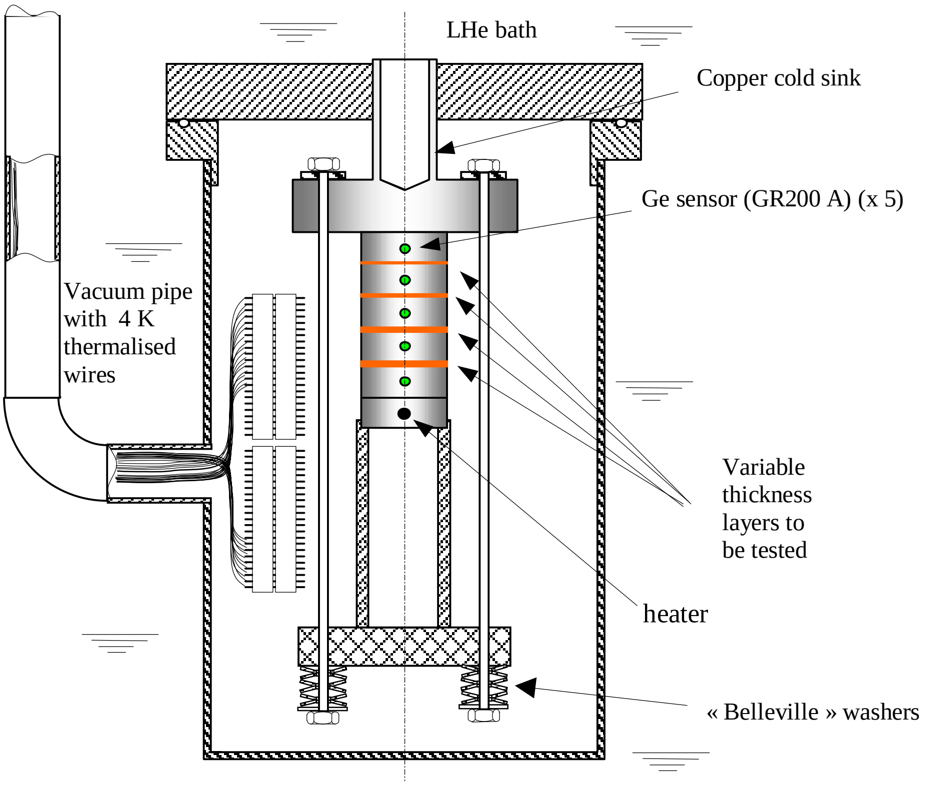

FIGURE 1. Experimental apparatus.

Samples are made with several layers of the product to be tested, separated by blocks of 1050 Aluminium alloy 10 mm thick. Temperature sensors are located in the middle of this blocks. Sensitive elements used are Germanium resistance sensor XGR-200A-3000-2 from Lake Shore Cryotronics. Temperature difference inside aluminium blocks is small enough to be negligible in comparison with the one in the layer. Material thickness is measured with a precision of 0.02 mm.

MEASUREMENTS ON THE PRE-IMPREGNATED FIBER-GLASS EPOXY COMPOSITE

Each composite sample is made with 5 blocks of aluminium separated with several layers of pre-impregnated tape IVA® (97 33/2)[5] (0.32 mm thick per layer). Thickness of composite was varied to determine thermal boundary resistance. Aluminium blocks are first sand-blasted and degreased by an ultrasonic method in alcohol, braised, then assembled with the pre-preg. Composite is afterwards polymerized in a vacuum oven (thermally cycled at 80°C for 4 h and at 120°C for 5 h). Insulation thicknesses between 5 blocks are about 0.26, 0.95, 1.9 and 2.8 mm. Three samples have been realized. Figure 2 shows the typical cross-section and picture of these samples.

The tests have been carried out on three multi-layer samples (#1,2,3) with 6 different heat fluxes (up to 50 mW), the smallest composite thickness being located on the cold sink side, except for the sample 1 which has been also tested in the opposite position (results 1a and 1b respectively).

Figure 3 presents the results, which are very consistent and show a low noise. The largest error bars observed correspond both to the thinner layer (for which the error on the thickness measurements reaches 10%) and to the smallest heat fluxes, where temperature differences are close to the temperature sensors incertitude. The thermal conductivity measured at 4.5 K is about 50 mW/m.K, which is much lower than that of G10 [6] initially used for our thermal calculations. Our results confirm measurements obtained by LASA [7].

Thermal boundary resistance ($R_h = \Delta T.S/Q$) between 1050 Al alloy and the composite is estimated from our results with different thicknesses. Total thermal resistance R_{tot} for each layer is assumed to be the sum of thermal bulk resistance R_{bulk} and thermal boundary resistances R_h.

$$R_{tot} = R_{bulk} + 2 \cdot R_h \tag{4}$$

with

$$R_{bulk} = \frac{l \cdot (T_h - T_c)}{\int_{Tc}^{Th} \lambda(T).dT} \tag{5}$$

For a given temperature (T≈5.5 K), an extrapolation of the $R_{tot}(l)$ law at zero thickness (see figure 4) leads to find a very small value in comparison with bulk properties ($1/R_h > 2000$ W/m^2.K around 5 K, consistent with previous works [8]).

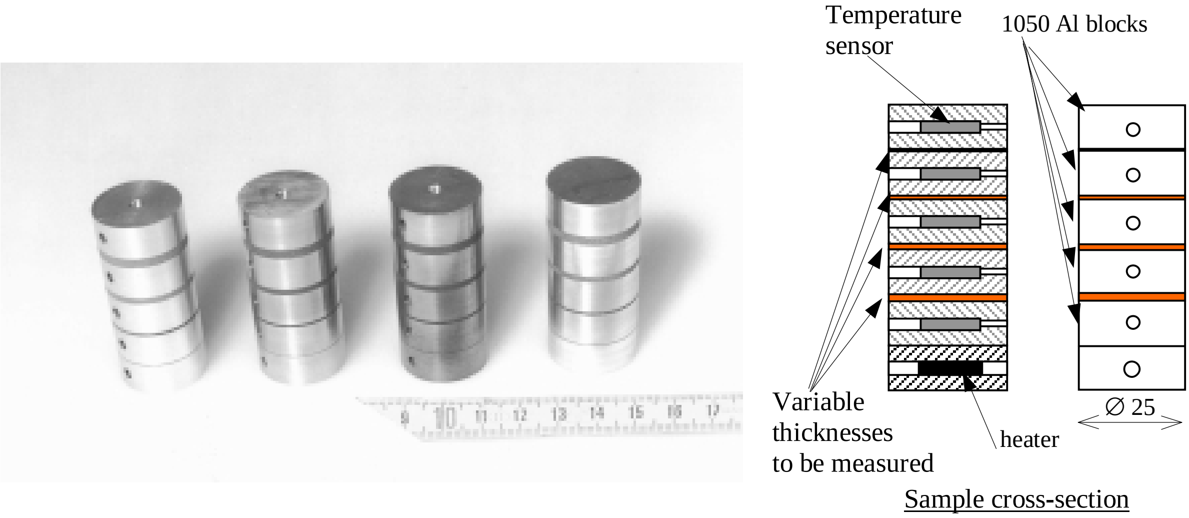

Figure 2 . Multi layers composite samples and cross-section scheme

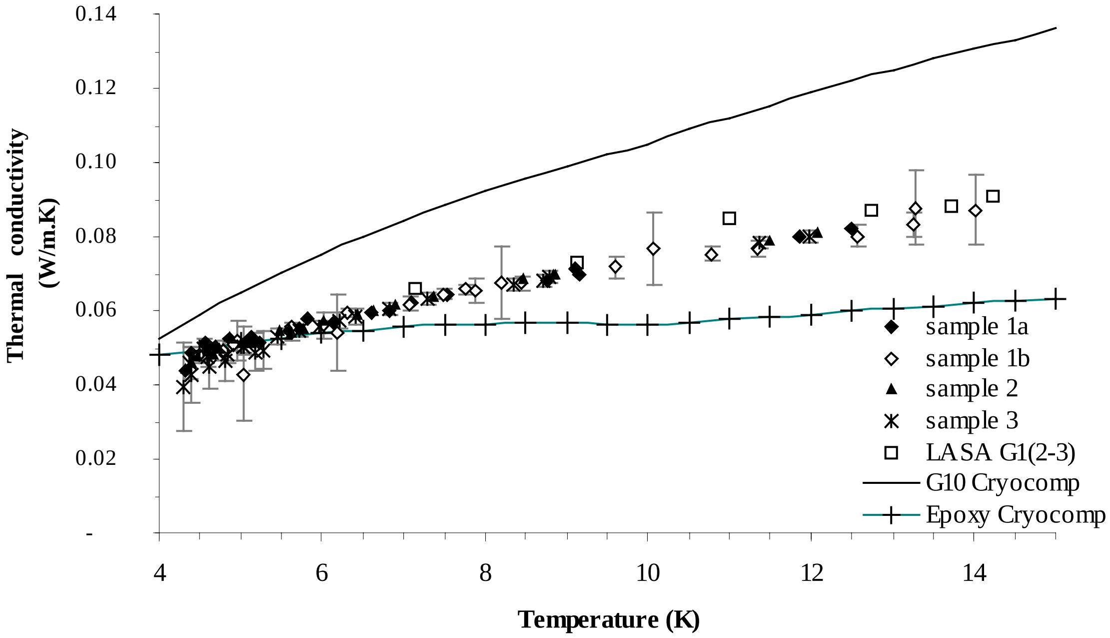

FIGURE 3. Thermal conductivity of pre-impregnated multi-layers composite

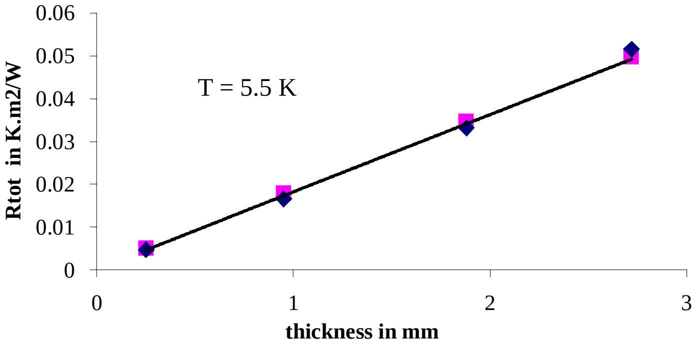

FIGURE 4. Estimation of the Al/composite thermal boundary resistance.

MEASUREMENTS ON VARIOUS BASED-EPOXY GLUES

Four epoxy glues have been measured at cryogenic temperatures using MECTI; i.e Eccobond 285 [9], Stycast 2850FT [10], Poxycomet F [11], DP 190 [12]. Eccobond and Stycast are traditionally used in many cryogenic assemblies where good mechanical behaviour and electrical insulation are needed. Their thermal conductivities at low temperature are known to be relevant to cryogenics contacts. To further its thermal conductivity, metal particles addition is employed as a solution. Unfortunately, this is unfavorable from the electrical insulation point of view. Poxycomet series is one of such glues (up to 80% Al) with a good mechanical behavior in aluminium assembly at low temperatures but with an unknown thermal conductivity. DP190 is often used in low temperature assembly [13] and it combines electrical insulation (up to 30 kV/mm) with good mechanical properties at low temperatures (remains sufficiently “soft” at low temperatures). Its thermal conductivity is also unknown in this range of temperature.

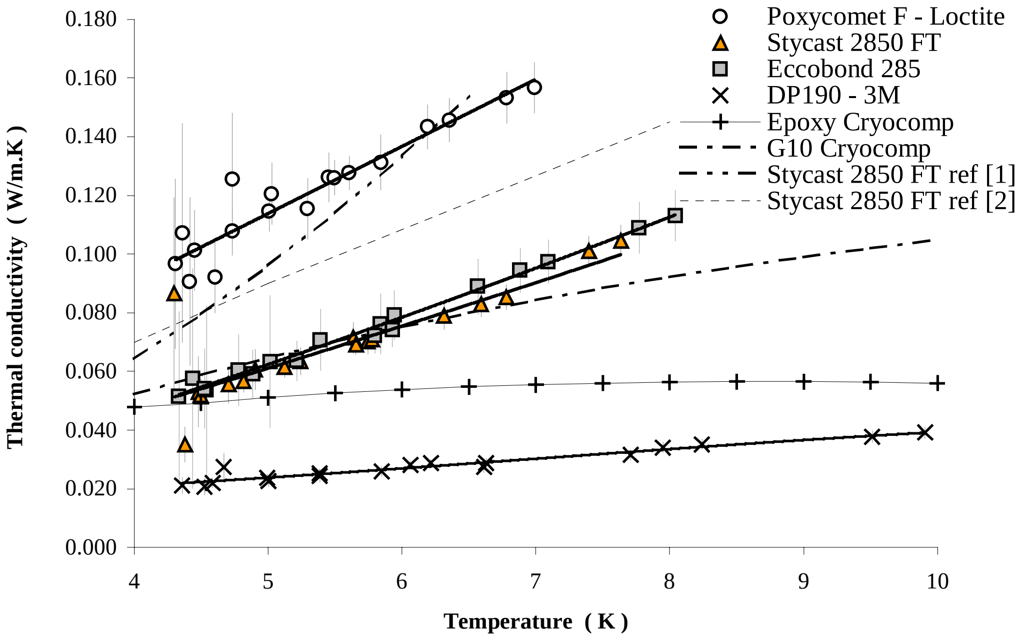

FIGURE 5. Thermal conductivity of epoxy-based glues.

Using 2 samples for each glue with 4 aluminium blocks and 3 layers of equal thickness (e ≈ 0.7 mm), thermal conductivity has been measured. Results are shown in figure 5. Near 4 K, Stycast 2850 FT preparation (with Catalyst 9) follows a $\lambda(T)=a.T^n$ law with a ≈ 0.0095 and n ≈1.2, closer than the quasi similar linear behavior already observed with this catalyst [2] but in contrast to 0.0053/1.8 found in another work (with catalyst 11) [1]. All over the range 4-8 K, thermal conductivity values are 30 % lower than these works. In this range, λ(T) of Eccobond 285 (with catalyst LV24) behaves very similarly to that of Stycast. DP190 has poor and rather constant thermal conductivity, between 0.02 and 0.03 W/m.K. This value is lower than pure epoxy resin, probably because it contains 50% hardener. As presumed with its high charge in aluminium, (≈80% in mass), Poxycomet F leads to twice as much thermal performance as Stycast 2850 FT and gives also a linear law in this range as 0.023T. The increase in thermal performance is finally poor for this so-called “pasty aluminium”.

CONCLUSIONS

Pre-impregnated epoxy-fiberglass composite measurements show a significant difference in its thermal conductivity from the values of G10 found in literature [4], i.e from 16 to 30 % less between 4 and 10 K. When values of G10 are used in simulation calculations instead of them of pre-impregnated composite, this difference must be taken into account. Thermal boundary resistance between the composite and Al has been evaluated at less than 5.10^{-4} $K.m^2/W$ which can be considered negligible in our analysis.

Bonding glues measurements give additional data to be used in low temperature assemblies. A thermal conductivity range from 0.02 W/m.K up to 0.1 W/m.K at 4 K has been measured for the 4 commercial epoxy- based glues.

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