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Thermal conductivity and Kapitza resistance of cyanate ester epoxy mix and tri-functional epoxy electrical insulations at superfluid helium temperature

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Abstract

In the framework of the European Union FP7 project EuCARD, two composite insulation systems made of cyanate ester epoxy mix and tri-functional epoxy (TGPAP-DETDA) with S-glass fiber have been thermally tested as possible candidates to be the electrical insulation of 13 T Nb_3Sn high field magnets under development for this program. Since it is expected to be operated in pressurized superfluid helium at 1.9 K and 1 atm, the thermal conductivity and the Kapitza resistance are the most important input parameters for the thermal design of this type of magnet and have been determined in this study. For determining these thermal properties, three sheets of each material with different thicknesses varying from 245 μm to 598 μm have been tested in steady-state condition in the temperature range of 1.6 K - 2.0 K. The thermal conductivity for the tri-functional epoxy (TGPAP-DETDA) epoxy resin insulation is found to be k=[(34.2±5.5).T-(16.4±8.2)]×10^{-3} $Wm^{-1}K^{-1}$ and for the cyanate ester epoxy k=[(26.8±4.8).T-(9.6±5.2)]×10^{-3} $Wm^{-1}K^{-1}$. For the Kapitza resistance, R_k, the best curve fitting the experimental data is described by R_k=(3057±593)x10^{-6}.$T^{(-1.79\pm0.34)}$ m^2KW^{-1} for the TGPAP-DETDA insulation and R_k=(4114±971)x10^{-6}.$T^{(-1.73\pm0.41)}$ m^2KW^{-1} for the cyanate ester epoxy insulation. Our results are compared with other epoxy based composite electrical insulation found in the literature.

1. Introduction

Within the framework of the FP7 European project EuCARD, an Nb_3Sn magnet is under development to serve as a test bed for future high field magnets and to upgrade the vertical CERN cable test facility [1]. The magnet is designed to achieve a magnetic field of 13 T at 1.9 K. As opposed to Nb-Ti magnets, Nb_3Sn magnets require the use of fiber-based composite electrical insulation that can resist the heat treatment that creates the intermetallic Nb_3Sn component. The fiber is impregnated with an epoxy resin polymer after the heat treatment step. This type of insulation is called "dry" insulation, *i.e.* impermeable to liquid helium. One of the major inconveniences for this type of magnet is that the coolant does not have an intimate contact with superconducting cables therefore reducing the cooling capacity of such magnets. Heat generated in superconducting cables during the AC losses due to field ramp rate, quench process, beam losses and synchrotron radiation has to be removed from the superconducting cables to the cold source via the surrounding electrical insulation which is in fact the main thermal barrier. This process is governed by two mechanisms: the thermal conductivity through the insulation itself and the Kapitza resistance at the helium-solid boundary. These thermal properties can be obtained only by experiments. The present paper concerns the experimental results of the thermal characteristic of the cyanante ester epoxy mix and tri-functional epoxy (TGPAP-DETDA) developed during the EuCARD project [1]. The principle of our experiment is based on the measurement of the overall thermal resistance of the tested sheets separating two isothermal superfluid helium baths, where one bath is heated and the other is temperature controlled [2, 3]. We measured temperature differences across the sheets as a function of heat flux. For extracting the Kapitza resistance and the thermal conductivity as a function of temperature, three thicknesses of each material are tested in the temperature range of 1.6 K - 2.0 K.

2. Samples preparation

The composite insulation materials used during measurements were produced using a vacuum impregnation process, which is similar to vacuum impregnation of a magnet structure. A stack of glass fiber was placed in a tray and aluminium plates placed in between the layers to create individual laminates. This was then evacuated to better than 0.1 mbar pressure in a vacuum tank at a temperature of 40°C. Liquid polymer was prepared by mixing and degassing to a similar pressure in a separate vacuum chamber, before being introduced to the glass fiber stack. After submerging the glass fibre with polymer, the pressure was returned to atmospheric pressure to impregnate the glass fibres and the stack was moved to a heated press where it was cured under 0.1 MPa pressure. The cured laminates were then removed from the tray using the aluminium plates to easily separate them. For the cyanate ester-epoxy blend (by weight 60% epoxy and 40% cyanate ester), a cure cycle of 6 hours at 100°C, 4 hours at 120°C and 17 hours at 150°C was used. For the tri-functional epoxy TGPAP-DETDA, a cure cycle of 14 hours at 70°C and 15 hours at 90°C was used. The samples have a diameter of 100 mm and are simply cut from the sheets produced by the process described above.

3. Experimental apparatus

To measure the temperature difference across the samples, the “drum” technique is used [2, 3]. The experimental apparatus, showed in Fig. 1, is composed of five stainless steel flanges. The sample sheets are located on both sides of a central cylindrical support. To prevent the flow of superfluid helium from the inner bath to the external helium reservoir, the sample sheets are glued to the flanges with epoxy resin, which creates an approximately effective diameter of heat transfer of 80 mm (cf. Fig 2). Flanges are screwed to the central support instrumented by a Cernox temperature sensor and a heater having an electrical resistance of 8 Ω. The apparatus is placed in a pressurized superfluid helium bath of a “Claudet” type cryostat [4].

The superfluid helium fills the internal volume, which is considered isothermal in superfluid helium, via the 0.4 m long capillary tube, wrapped and glued around the stainless steel central cylindrical support. The capillary tube also carries the instrumentation wires for the temperature sensors and heater. In that way it reduces the heat transfer cross section to 0.153 mm^2 and therefore the heat losses through the capillary. Before each cool down, the experimental set-up is purged three times where air is replaced by helium gas by pumping for several hours. In that way, the air inside the inner volume is replaced by helium gas through the capillary.

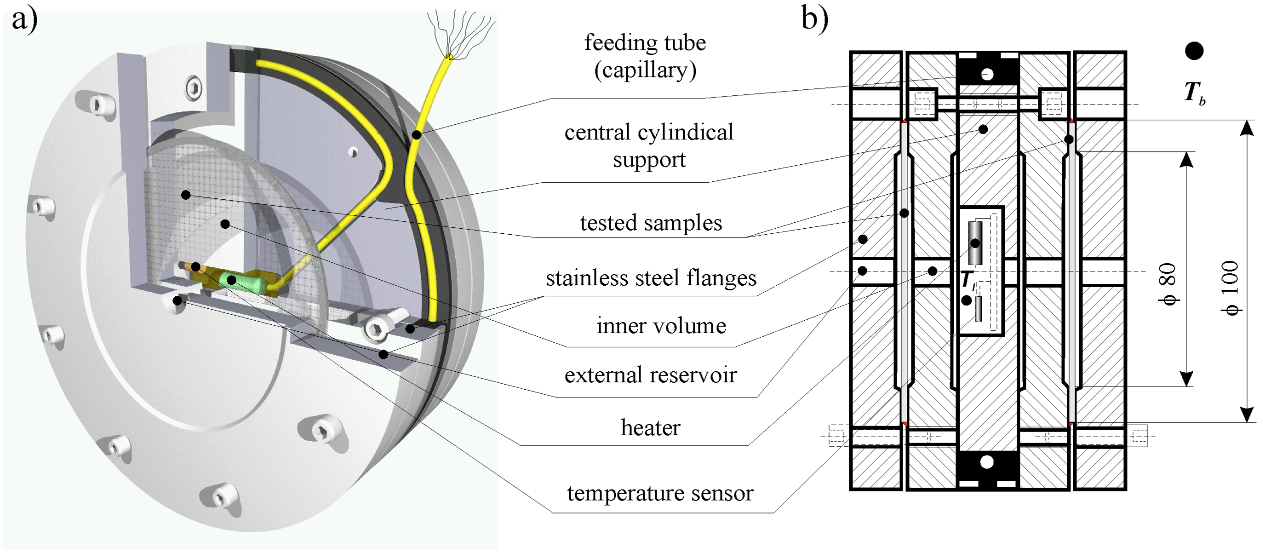

Fig. 1. Schematic of the experimental set-up

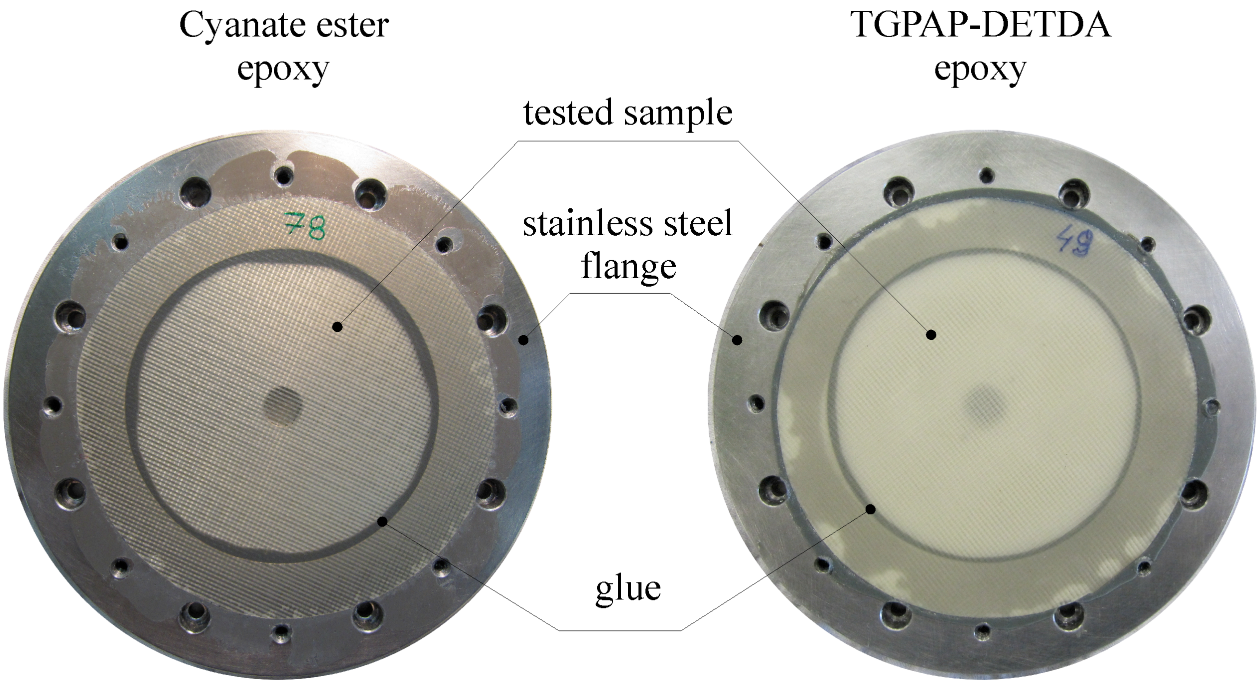

Fig. 2. Pictures of two sample sheets glued on their support flange

4. Theoretical background

When a heat flux Q_s is applied perpendicularly to the samples in the inner bath, a temperature difference $\Delta T=T_i-T_b$ is created across the tested sheets and between the internal T_i and the external bath T_b. The value of the temperature difference during steady state heat load depends on the thermal resistance of the insulation R_S due to the conduction and the Kapitza resistance $R_{Kapitza}$ between the solid and superfluid helium [2,3]. According to the Khalatnikow's theory [5], the heat flux between a solid and a liquid can be calculated based on the acoustic mismatch theory and the Kapitza resistance has a T^3 temperature dependence. It has been experimentally proven that the value of the Kapitza resistance is not only a function of the temperature but also depends on the solid material properties, the solid surface conditions, pressure, etc [6]. Therefore, in practice the exponent of the temperature dependency varies around 3 from 2.5 to 4.5 [7]. Also as mentioned in [7], the exponent is temperature dependent and increases with temperature especially near T_λ which is a consequence of direct photon or roton emission, and a much shorter phonon wavelength as well.

According to the mismatch theory [5], a heat flux between the surface of the solid at T_s and the He II at a temperature of T_{HeII} can be expressed by

$$\frac{Q_S}{A_S} = h_K(T_s^n - T_{HeII}^n), \qquad (1)$$

where Q_s is the heat flux going through the solid, A_s is the effective area of the heat transfer and h_K is the Kapitza conductance.

In our experimental set-up, the heat generated in the inner bath goes through the inner sample surface, the sample and the outer sample surface facing the cryostat bath. This can be formulated as

$$\frac{Q_S}{A_S} = h_K(T_i^n - T_1^n) = \frac{k}{l}(T_1 - T_2) = h_K(T_2^n - T_b^n) \qquad (2)$$

where T_i, T_1, T_2 and T_b are, respectively, the temperatures of the inner helium volume, the surface of the sample from the inner volume side, the surface of the sample from the cryostat bath side and the cryostat bath. k and l are the thermal conductivity of the tested material and its thickness.

If the temperature rise ΔT during the experiment is much smaller than the temperature of the bath we can assume that $T_i \approx T_b$, then after a linearization described in details in [2, 3], Eq. (2) is simplified to

$$\frac{Q_S}{A_S} = n\,T_i^{n-1}\,h_K(T_i - T_1) = \frac{k}{l}(T_1 - T_2) = n\,T_i^{n-1}\,h_K(T_2 - T_b), \qquad (3)$$

which leads to the expression of the overall thermal resistance of the sample, R_s, as a function of the bath temperature, T_b

$$R_s = A_s\,\frac{\Delta T}{Q_S} \approx \frac{2}{n\,T_b^{n-1}\,h_k} + \frac{l}{k}. \qquad (4)$$

Summarizing, the first term on the right side of Eq. (4) can be identified as two Kapitza resistances R_K, the second term is the conduction resistance R_λ of the sample, *i.e.*

$$R_s = 2.R_k + R_\lambda. \qquad (5)$$

The overall thermal resistance of the insulation can be obtained from equation (4) only when the temperature difference ΔT during one series of data points has a linear evolution with Q_s *i.e.* R_s is constant. For our experimental measurements, this condition is usually satisfied above a ΔT of 10 mK as the Figure 3 shows, where the R_s evolution for the 288.8 μm thick TGPAP-DETDA and the 245.1 μm thick cyanate ester mix samples as a function of ΔT is depicted at a bath temperature of 1.7 K. It is a typical result and such curve is found at different bath temperatures. In an experiment the total heat dissipated by the electrical resistor in the inner bath is mainly transferred through the samples but some amount goes through the helium in the capillary and the stainless steel flanges. At low values of heat dissipation showed in Figure 3, a nonlinear part is observed, where the heat is transferred mainly through the helium in the capillary.

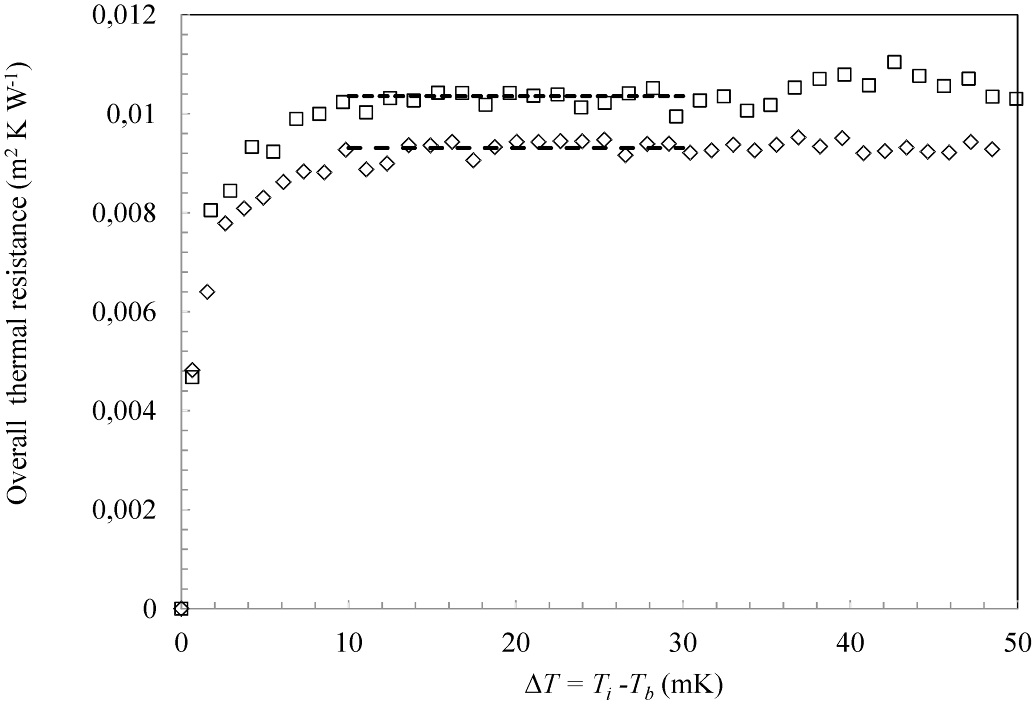

Fig. 3. Total thermal resistance of the 288.8 μm thick TGPAP-DETDA ◊ and 245.1 μm thick cyanate ester mix □ samples as a function of temperature difference at 1.7 K bath temperature.

Above a ΔT of 10 mK, it can be noticed that the value of the total resistance is quasi constant and varies around an average value (shown in Figure 3 as a dotted lines). As shown in [2, 3], the amount of the heat transferred by superfluid helium through the capillary and the stainless steel flanges can reach 20% of the dissipated heat for small ΔT around 1 mK and only a few percent for ΔT comprised between 10 mK and 30 mK. To reduce the systematic error due to the heat loss through the capillary, the thermal resistance will be determined in that ΔT range, for each sample and bath temperature.

5. Measurement Errors

The dissipated heat is controlled and measured by a Keithley 2400 source meter, which is the combination of a power source and a voltmeter, and with an uncertainty that does not exceed 1% of the controlled value. The uncertainty comes mainly from the measurement of the voltage across the heater since the current is controlled with high stability. To measure the temperature in the inner volume a Cernox temperature sensor, with a four-wire technique and a DC battery current source with value of 1μA ±0.05%, are used. The voltage drop across the temperature sensor was measured by a NI SCXI voltage measurement system. For the inner volume temperature, T_i, the experimental standard variation of the resistance measurement was lower than ±0.5 Ω which corresponds to a temperature variation from ± 30 μK at T_b = 1.6 K to ± 70 μK at 2.0 K. The calibration processes was repeated after every warming and cool down processes. The voltage measurement, the current value uncertainty and the propagation of errors through the calibration curve lead to a temperature error of ± 0.2 mK. The temperature of the cryostat bath T_b is controlled with a precision of 1 mK (and an accuracy of 5 mK) by a LakeShore 332 temperature controller with a Cernox thermometer and the 8 Ω heater.

The thickness of every sample was measured, five times, at four points with the accuracy of ± 1 μm and the average value was considered as a representative thickness. Before and after mounting the sample in the experimental apparatus, the sample was cleaned using methyl alcohol. To determinate the real values of the effective heat transfer area A_s, a digitized picture and pixel counting software was used. Firstly the contour of the effective area is defined thanks to the difference in pixel intensity between the glued and unglued part of the sample. Secondly, the software counts the number of pixels within the unglued part. Knowing the number of pixels for the total surface of the flange (80 mm in diameter), the effective area can be deduced. This method gives precision of 0.2 % for A_s. The geometrical specifications of the tested samples are summarized in Table 1.

6. Fitting procedure

In order to simultaneously extract the thermal conductivity and the Kapitza resistance as a function of temperature, different thicknesses of the same material at different bath temperature must be tested. The first step is to determine the overall thermal resistance R_s, by measuring the total temperature difference, ΔT, against the heat flux through the sample Q_s.

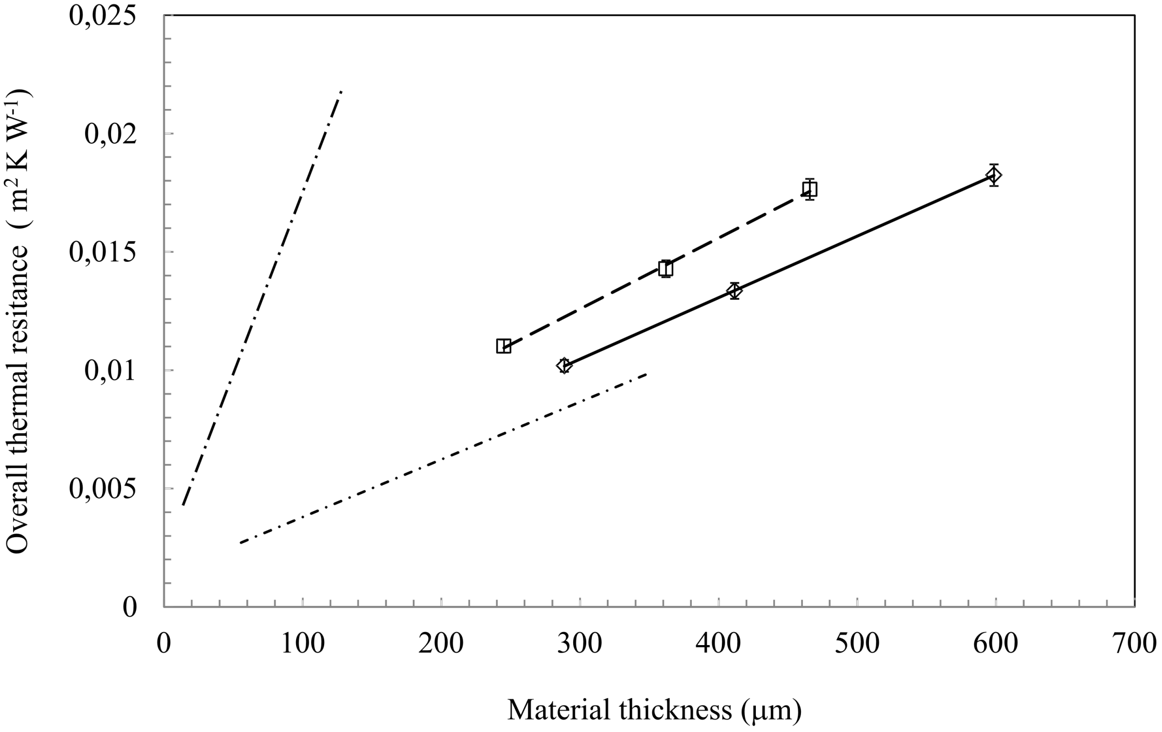

Fig. 4. Overall thermal resistance R_s of tested samples, TGPAP – DETDA ◊, cyanate ester mix □, and Kapton [2] —·—·—, epoxy resin fiberglass tape [3] -·-·- as a function of thickness at 1.7 K.

For each bath temperature, the thermal resistance is plotted as a function of the thickness and the data are fitted to Eq. (4) using a non-linear least squares method where the independent variable is the thickness, l and the dependent variable is the thermal resistance R_S. The typical results for the 245.1 μm thick cyanate ester mix sample and the 288.8 μm thick TGPAP-DETD at 1.7 K are presented in Figure 4. The inverse of the slope gives the thermal conductivity k and the y-intercept represents two Kapitza resistances (according to Eq. (5)). We compared the experimental data with electrical insulations such as Kapton [2] and epoxy resin fiberglass tape [3] obtained using the same "drum" method. For all cases it is observed that the evolution of the overall thermal resistance has a linear trend. It can be remarked that the main influence on the overall thermal resistance is the conduction thermal resistance R_λ, which is around seven times larger than the Kapitza resistance R_K. Cyanate ester epoxy mix, tri-functional epoxy and epoxy resin fiberglass tape are characterized by almost the same value of overall thermal resistance; the maximum difference is around 15 %. In comparison with Kapton, the current insulation material of Nb-Ti accelerator magnets, the difference in overall thermal resistance is more than five times lower.

7. Results and discussion

6.1. Thermal conductivity

The results of the thermal conductivity as a function of the temperature are shown in Figure 5. The tested materials are characterized by amorphous structure. For those materials the thermal conductivity below 1 K is described by a quadratic dependency with temperature, for higher range (5 K - 15 K) a plateau is reached [3]. In the range between 1-5 K the thermal conductivity have a temperature dependence of T^n where n is in the range from 0 to 2. From our measurement, in the investigated temperature range, it can be observed that thermal conductivity of the tested materials is proportional to the temperature i.e. $n=1$, which agrees very well with the results obtained for similar materials such as epoxy fiberglass tape [3]. The thermal conductivity of both materials is given by the following expression,

$$k=[(34.2\pm5.5).T-(16.4\pm8.2)]\times10^{-3}\ \mathrm{Wm^{-1}K^{-1}} \qquad (6)$$

for the TGPAP-DETDA sample and

$$k=[(26.8\pm4.8).T-(9.6\pm5.2)]\times10^{-3}\ \mathrm{Wm^{-1}K^{-1}} \qquad (7)$$

for cyanate ester epoxy mix sample.

We have compared the thermal conductivity of the tested materials with other insulations such as epoxy [8], epoxy resin fiberglass tape [3] and Kapton [2] in Figure 5. The TGPAP-DETDA epoxy insulation has the largest thermal conductivity, which in comparison to Kapton is characterized by six fold higher value. From the Figure 5, it can be observed that for the tested insulations and epoxy resin fiberglass tape, the temperature evolutions are parallel to each other. These insulations are fabricated in the very similar way and consist of approximately 50% volume fraction of fiberglass tape, explaining the similarity of the temperature dependency. The small difference in the value of thermal conductivity could be a consequence of the use of a different type of resin. To the best of our knowledge, none of thermal conductivity of the raw material constituting the tested insulations has been presented in the literature. Thus it is difficult to quantify the influence of each component on the thermal conductivity of these composite materials.

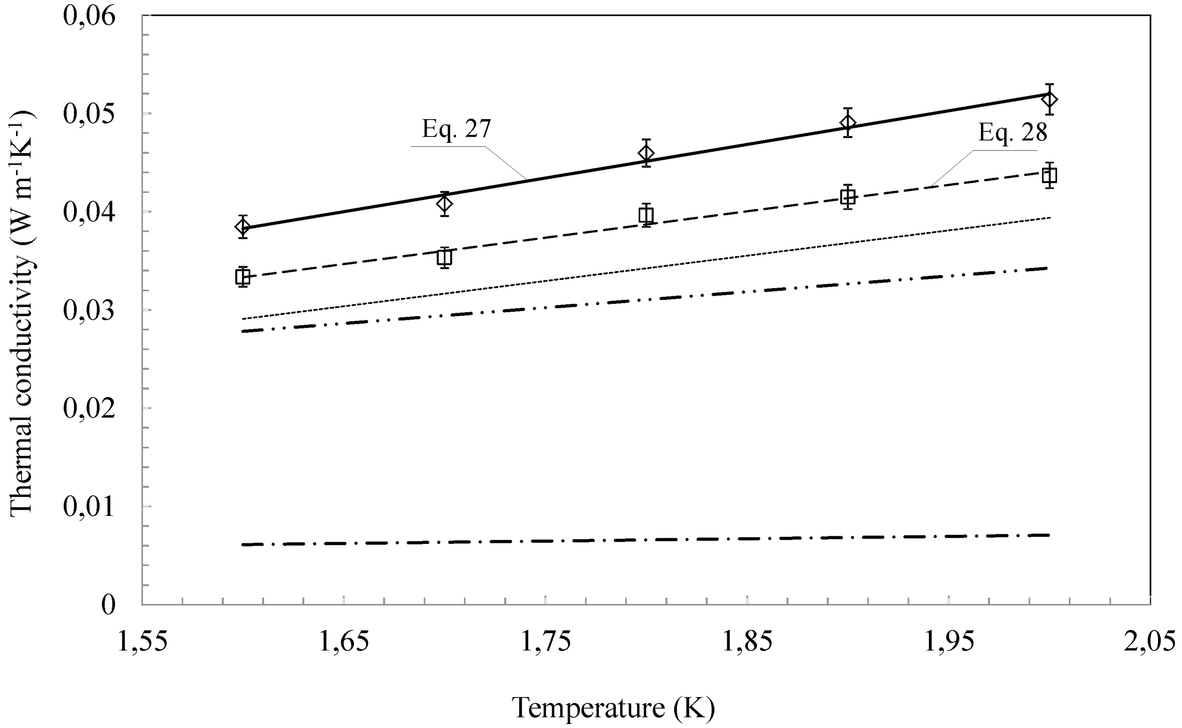

Fig. 5. Evolution of the thermal conductivity with temperature: TGPAP – DETDA ◊, cyanate ester mix □, and Kapton [2] —·—·—, epoxy resin fiberglass tape [3] · · · and epoxy [8] -··-··-.

6.2 Kapitza resistance

Figure 6 presents the data obtained during measurements and the best fit to the Kapitza resistance results. For our experiment, the data can be expressed by the following expressions for the TGPAP-DETDA epoxy,

$$R_k=(3057\pm593)\text{x}10^{-6}.T^{(-1.79\pm0.34)},\ \text{m}^2\text{KW}^{-1} \qquad (8)$$

and for the cyanate ester epoxy mix,

$$R_k=(4114\pm971)\text{x}10^{-6}.T^{(-1.73\pm0.41)}\ \text{m}^2\text{KW}^{-1}. \qquad (9)$$

The observed non-linear law of the Kapitza resistance (for TGPAP-DETDA $R_k \propto T^{-1.79}$ and for cyanate ester epoxy mix $R_k \propto T^{-1.73}$) is consistent with the theory discussed above and according to the first term of Eq. (4), the heat transfer coefficient value for TGPAP-DETDA is equal to h_k=235±47 Wm^{-2}K$^{-2.79}$ and for cyanate ester epoxy h_k=178±12 Wm^{-2}K$^{-2.73}$.

The data can be also fitted based on the acoustic mismatch theory [4] which assumed that n=3. For TGPAP-DETDA epoxy the Kapitza resistance is

$$R_k=(5971\pm779)\text{x}10^{-6}.T^{-3}\ \ \text{m}^2\text{KW}^{-1}, \tag{10}$$

and for cyanate ester epoxy mix, it is

$$R_k=(8253\pm1129)\text{x}10^{-6}.T^{-3}\ \ \text{m}^2\text{KW}^{-1}. \tag{11}$$

Equations (10) and (11) are depicted as dotted lines in Figure 6. The heat transfer coefficients associate with Eq. (10) and Eq. (11) for TGPAP-DETDA is h_k=112±21 $\text{Wm}^{-2}\text{K}^{-4}$ and for the cyanate ester epoxy mix insulation h_k=61±7 $\text{Wm}^{-2}\text{K}^{-4}$.

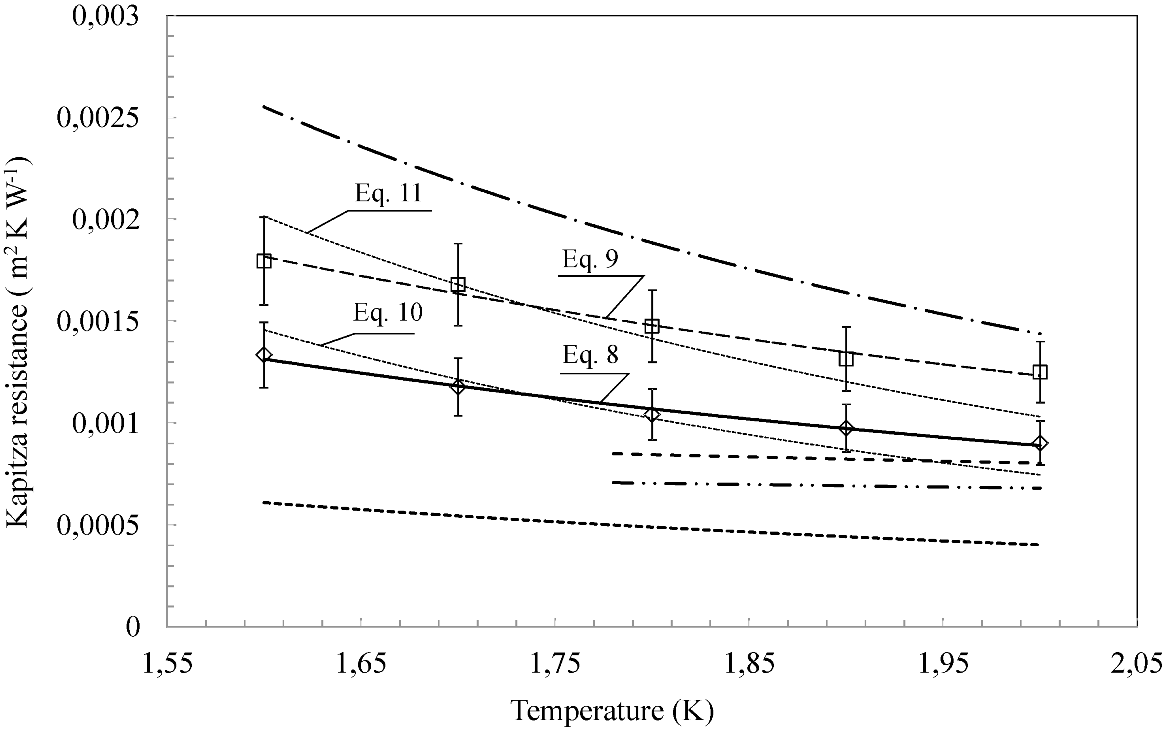

Fig. 6. Evolution of the Kapitza resistance with the temperature: TGPAP – DETDA ◊, cyanate ester mix □, and Kapton [2] —·—·—, epoxy resin fiberglass tape [3] —, Stycast coating on polished surface [9] -··-··-, Stycast coating on oxidized surface [9] - - -.

In Figure 6 we compared the experimental results of Kapitza resistance for different materials such as: Kapton [2], epoxy resin fiberglass tape [3] and Stycast coated on polished and oxidized surfaces [9]. The obtained results are in the same order of magnitude than the insulations found in the literature and presented in Figure 6. Our result is almost two times higher than the one obtained for epoxy resin fiberglass tape [3] even though the external surfaces of the insulations are totally covered by the resin. We can assume that the Kapitza resistance is directly related to the epoxy resin only. We think that the difference is not only a consequence of the different resin but also results from the different surface conditions [6, 7]. The epoxy resin fiberglass tape was characterized by an "egg tray" shape surface while our tested sample the surface has a smooth and flat surface [3]. A very similar surface condition was tested by Iwamoto on Stycast resin [9] and our results are in good agreement with his data; the difference is in the range of 20% - 30%.

8. Conclusions

Thermal conductivity and Kapitza resistance of cyanate ester epoxy mix and a tri-functional epoxy (TGPAP-DETDA) - two potential insulation materials for the Nb_3Sn high field magnet under development in the HFM EuCARD project have been measured at pressurized He II temperatures. The study shows that the thermal resistances of the tested electrical insulations are very similar to fiberglass tape impregnated with resin since they have the same composition of 50% fiberglass tape and 50% resin but are five times lower than for Kapton. In the investigated range of the temperature, it was found that the value of the thermal conductivity increases linearly with temperature. The obtained value of the Kapitza resistance is very similar to the data published in the literature. Moreover, the thermal characterization of these electrical systems at higher temperature is required for the thermal design of the magnet and additional measurement such as thermal conductivity and heat capacity are foreseen between 4 K and 300 K.

Acknowledgements

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