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THE THERMAL PROPERTIES OF COMPOSITE MATERIALS

Final Technical Report

by H. M. Rosenberg

January 1978

EUROPEAN RESEARCH OFFICE United States Army London, England



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GRANT NUMBER DA-ERO 75-G-018

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UNCLASSIFIED **R&D 1651** SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) READ INSTRUCTIONS **REPORT DOCUMENTATION PAGE** BEFORE COMPLETING FORM 2. GOVT ACCESSION NO. 3. RECIPIENT'S CATALOG NUMBER 1. REPORT NUMBER TITLE (and Subtitle) FINAL TECHNICAL REPO г, THE THERMAL PROPERTIES OF COMPOSITE MATERIALS JAN 75 - 29 JAN 78 AUTHORA . CONTRACT OR GRANT NUMBER(+) H. M. ROSENBERG DAERO-75-G018 10. PROGRAM TE MENT, PROJECT, TASK PERFORMING ORGANIZATION NAME AND ADDRESS CLARENDON LABORATORY 6.11.024 T161102B32D-00-UNIVERSITY OF OXFORD -OXFORD 428 11. CONTROLLING OFFICE NAME AND ADDRESS 12. REDOR U.S. ARMY R&S GROUP (EUROPE) JANNA 78 **BOX 65** FPO 09510 29 15. SECURITY CLASS. (of this report) 14. MONITORING AGENCY NAME & ADDRESS(If different from Controlling Office) UNCLASSIFIED 15e. DECLASSIFICATION/DOWNGRADING SCHEDULE 16. DISTRIBUTION STATEMENT (of this Report) DI Approved for Public Distribution Unlimited Release. 17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, If different from Report) 5) DA-ER0-75-6-028 18. SUPPLEMENTARY NOTES 19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Thermal Expansion . Themal Conductivity Departments, establishments or contractors the anove vish . 1a Metal-Epoxy Composites Low Temperature Properties 2 20. ABSTRACT (Continue on reverse side if necessary and identify by block number) "see over" DD 1 141 73 1473 EDITION OF I NOV 65 IS OBSOLE SSIFIED Data Entered) ICATION OF THIS PAGE (S. M. A. Cours

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The thermal expansion of carbon-fibre/epoxy-resin composites has been measured from 20 to 440 K for specimens in which the fibres were unidirectional and also for those in which they were laid in layers crossed at 90° to one another. The crossed fibre specimens show a very high dimensional stability on cooling and for one specimen there was no detectable change in length between 20 and 250 K.

The thermal expansion and conductivity of a sample of a Mg matrix/ 35% carbon-fibre composite have been measured. The thermal expansion was measured from 20 to 460 K. The expansion above room temperature is very small whilst from 20 to 250 K there is virtually no dimensional change at all to within a few parts in 10^5 . The thermal conductivity is reasonably high and is roughly that due to the magnesium content of the composite.

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Abstract

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Introduction

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This report contains accounts of three investigations which have been undertaken on the thermal properties of composite materials. These are as follows:

- An investigation of the thermal expansion of epoxy-resin/particle composites from 77 to 450 K in which a particle size-dependent effect has been observed in the region of the glass-plastic transition of the epoxy-resin matrix.
- (2) A study of the thermal expansion of carbon-fibre/epoxy-resin composites which indicate that these materials show extremely high dimensional stability both at room temperature and also at low temperatures.
- (3) A preliminary investigation of the thermal conductivity and expansion of a metal-matrix composite - magnesium + 35% carbonfibre. This material has a very low thermal expansion and yet it is a good heat and electrical conductor.

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Chapter 1

The Thermal Expansion of Epoxy-Resin/

Particle Composites - a Size Effect

by M. de F. F. Pinheiro and H. M. Rosenberg

1.1 Introduction

This paper describes measurements taken from 77 K to 450 K of the thermal expansion of composite materials made of an epoxy resin matrix which contain spherical particles of glass and copper. Our investigation stems from the work of Grubb (1975) who observed that above $360 \text{ K} (90^{\circ}\text{C})$ the thermal expansion of such composites appeared to be dependent on the size of the filler particles.

Many formulae have been proposed for the calculation of the thermal expansion coefficient of composite materials. Several authors have shown that the empirical rule of mixtures, which assumes that the expansion itself is additive on a volume basis, is not followed in several systems and that the thermally induced stresses between the phases due to differences in their thermal expansivities need to be taken into account (e.g. see the review article by Holliday and Robinson, 1973).

When a composite is allowed to cool from its preparation temperature down to room temperature its constituents will shrink. If the shrinkage of the matrix is higher than that of the filler (as is the case for an epoxy matrix) then the filler will be compressed by the matrix, and the latter may, in time relax, so as to relieve the stress. If the composite is heated again, the matrix tends to expand more than the particle, but its expansion will be restrained by adhesion at the interface. The thermal expansion of the whole sample will depend on both how effective the adhesion is to be able to sustain these thermally induced strains and also on the mechanical properties of the constituents.

Many of the expressions which have been proposed to take account of the mechanical interaction between phases are discussed by Holliday and Robinson^{*} (1973) and none of them predicts any effect which is dependent on the size of the particles. The treatments all assume purely elastic behaviour and also a perfect adhesion between the different phases.

Depending on the composite system under investigation some formulae fit the experimental data better than others, but there is little to choose between several of them (Grubb, 1975). In general, quite reasonable agreement can be achieved between theory and experiment.

*Note that the expression they quote for the theory of Wang and Kwei is incorrect.

1.2 Experimental details

(a) Sample preparation

Both the copper and glass ballotini filling materials were spherical and are commercially available in several different sizes. The separation into fractions containing particles with a narrow size distribution was done using test sieves for particles with diameters larger than 30 μ m and with an Alpine cyclone separator for particles smaller than 30 μ m. The average particle size of the various samples was deduced from microscopic observation.

The epoxy matrix used was Shell Epikote 828, 100 parts by weight with 90 parts Epikure NMA hardener and 0.5 part BDMA accelerator. After the mixture was degassed and heated up to 40°C to reduce its viscosity, the required weight of filler was thoroughly mixed in and the mixture was degassed again in a vacuum oven. This helped to prevent the formation of voids. The mixture was poured into coated glass tubes for casting. All the samples had the same curing conditions. They were held for two hours at 100° C and then the temperature was raised at 1° C per minute to 200°C and was held there for four hours. The moulds containing the mixtures were rotated at 2 rpm in the oven to prevent the particles from settling. The proportions of filler and resin in each sample after it was prepared was deduced from a measurement of its average density. For the present experiments volume concentrations, Vf, of filler of 0.3 and 0.5 were used for the copper specimens and 0.3 for those containing glass. Seven sizes of copper particle were used 5-10, < 30, 40-60, 75-90, 90-106, 106-125 and 125-150 µm. The glass particle sizes were 3.5-5, 75-90 (one run), and 180-200 µm. The specimens were cylinders approximately 20 mm in diameter and 12 mm high.

(b) Apparatus and procedure

A capacitative method was used to determine the thermal expansion. The change in length of a specimen was made to vary the electrode gap in a 3-terminal capacity cell (White, 1961). The capacitance was measured with an ASL capacitance bridge which compared the capacity of the test cell with that of an external 10 pf standard. A change in capacitance of 1 part in 10^6 could be detected. Two capacitance cells were in fact used; one was mounted in a cryostat and was used in the low temperature to room temperature range and the other was heated in a thermostatically controlled oil bath for measurements from room temperature up to 450 K. Further details of the experimental technique are given by Grubb (1975).

1.3 The glass transition temperature

When an epoxy-resin is heated it undergoes a second-order phase transition at a temperature T_g , the glass transition temperature. Below T_g the material is brittle, whereas above this temperature it becomes rubbery. T_g may be determined by hardness and deflection tests and also by the fact that the coefficient of thermal expansion increases at T_g . It is thought that at the transition thermal excitation enables parts of the molecular chain to rotate about its bonds. For the resin used in these experiments, Epikote 828, the value of T_g from deflection tests is given by the manufacturers as between 398 and 408 K (125-135°C).

1.4 Results on the Cu-filled specimens

Most of the specimens were measured in the range 293-450 K but a few were also investigated from 77-293 K. After the first run up to 450 K, all the specimens showed a relative shrinkage of about 10^{-3} on cooling down, but after subsequent runs this shrinkage was less than 10^{-4} . Although the samples had all been cured above 450 K (at 473 K) this shrinkage might be due to a post-cure effect in which additional cross-linking of the polymer chains can occur during the slow heating (4h) of the specimen in the first experimental run. It could also be caused by water vapour being driven off from the matrix as it is known that epoxy-resins do absorb water vapour from the atmosphere. Although this shrinkage is an interesting observation it does not appear to be material to our main results.

Measurements from the initial runs for the set of $V_f = 0.3$ Cu-filled specimens are shown in figure 1. The results are plotted as the relative change in length $\Delta L/L$, compared with the original length at room temperature. For comparison the expansion of unfilled epoxy and pure copper are also shown. The effect of particle size is quite evident. The larger the particles, the greater is the thermal expansion at particular temperature. A similar effect is shown for the $V_f = 0.5$ specimens in figure 2.

1.5 The effect of different heating rates

Three more runs were taken (i.e. second to fourth heatings) on the $V_f = 0.3$ specimens containing the < 30 and 125-150 µm filler. In the second and fourth heating the warm-up rate was very slow - about 8 h to go from 290 to 450 K and for the third heating the rate was the same as for the first run, i.e. 4 h to cover the same range. The results for the large particles are shown in figure 3 in which it can be seen that there is very little difference between all the runs. For the small particles, however, the dependence on warm-up rate is quite marked (figure 4). The expansion is substantially less for runs 1 and 3 (fast rate) than for runs 2 and 4 (slow rate). It is also clear from this figure and from figures 1 and 2 that anomalous behaviour only starts to show beyond 360 K, which is about the glass transition temperature for the resin.

Since there was an interval of several months between runs 2 and 3 it was possible that the difference between them might be due to some ageing or moisture absorption process. To check this a further three runs were made with an interval of less than 48 h between each. The first was very slow - 9 to 10 h and the second and third very fast - 2 h. The results again show (figure 5) a similar dependence on heating rate as was observed in the initial runs.

From all these results we conclude that if small particle fillers are used the glass/plastic transition, tends to be suppressed. This results in a slowing down of the transition and a rise in the temperature at which it occurs.

We also took measurements from 293 down to 77 K for both the $V_f = 0.3$ and 0.5 specimens and confirmed that there was very little, if any particle size dependence in this ragne (figure 6).

To confirm that the size effect was three dimensional and also

that it was not an experimental artefact, a few measurements of the overall thermal expansion from room temperature to 400 K were taken by measuring the displacement of oil in a density bottle containing a specimen. Whilst the accuracy of this method cannot compare with the capacitative technique, the results were broadly in agreement with the capacity measurements.

1.6 Experiments on glass-filled specimens

A short set of experiments was made on specimens with $V_f = 0.3$ in which the fillers were glass ballotini of 3.5-5 and 180-200 μ m diameter. These, like the Cu-filled specimens showed a shrinkage of about 10^{-3} after the first run but thereafter no further shrinkage was observed. Once again a size effect is present (figure 7) although it is not so pronounced as for the Cu-filled specimens.

1.7 Comparison with theory

As stated in the introduction none of the many formulae that have been proposed for calculating the expansion of a composite incorporates any dependence on particle size. Nevertheless, since the samples containing the large diameter fillers appear to be 'well-behaved', we have used some of the expressions to see whether any of them fitted our results. All these expressions are quoted in the review paper by Holliday and Robinson (1973). For brevity we only present one set of comparisons - those for Cu-filled specimens with $V_f = 0.3$ and < 30 and 125-150 µm particles (figure 8). The various calculated curves show that, for the 125-150 µm samples, very good agreement is obtained above room temperature with the expressions of Kerner (K), Fahmy and Ragai (F-R) and Wang and Kwei (W-K). Below room temperature the agreement is not so satisfactory. The figure also shows that the results for the < 30 µm specimens do not agree with any of the theories and this underlines our assumption that it is these specimens which are showing anomalous behaviour and not those with large particles.

The three relations which fit the 125-150 μ m results seek in their different ways to match the thermal and the elastic stresses around a particle plus its encircling region of matrix with the general surroundings due to the remainder of the sample. In the F-R and W-K theories the filler and matrix are assumed to be perfectly elastic and the stresses drop off sufficiently rapidly around a particle so that there is no appreciable overlap of stresses due to neighbouring particles. It would appear that whilst these assumptions might be valid for the 125-150 μ m specimens (the particles are about 50 μ m apart) this is not so for the < 30 μ m samples (\sim 10 μ m apart).

For the glass-filled specimens the agreement with the theoretical expressions is not so satisfactory even for the largest particles (figure 9). The K, F-R and W-K curve is a reasonable fit up to about 350 K, but then it predicts too high an expansion. Since this is roughly the glass transition temperature one is tempted to suggest that there is still a size effect present for the 180-200 μ m ballotini, but it seems unlikely and we have not attempted to check this.

1.8 The origin of the size effect

From the results which we have presented it seems clear that in samples containing small-sized particles the onset of the glass/plastic transition is suppressed and so the increased thermal expansion which is characteristic of the plastic state is not able to set in until a higher temperature is reached. This is borne out by the general shape of all the curves which have been presented and also by the fact that below the glass transition temperature very little, if any, size effect is observed. The influence of the heating rate also points to some mechanism which modifies the interactions between different parts of the polymer. It might be possible that the close proximity of the particles or the stresses which they induce could inhibit the cooperative rotation of the molecular chains which occurs at the plastic transition. Clearly the dependence on the heating rate points to a mechanism which has a fairly long activation time and which might be susceptible to small perturbations of the activation energy. Another factor which might play a part is the possibility that the resin, particularly at the higher temperatures, might not behave elastically. This would certainly mean that the principle of superposition of stresses could not be assumed and hence the various theories would not be valid. It should also be noted that the effect is not so marked in the glassfilled specimens for which, due to lower values of the elastic moduli and the expansion coefficient of glass, the internal stresses and strains should be less.

All these ideas, however, are speculative. The only piece of corroborative evidence regarding the suppression of the glass/plastic transition that we have been able to find is by P. W. McMillan (private communication) who noted that in thin films of glass (about 10 μ m thick) the transition was inhibited. This is a similar distance to that between our fine Cu particles and whilst the details of the observation are quite different from our experiments, it does again point to a suppression of glass/plastic transition when the distances involved are short.

1.9 Acknowledgements

We should like to thank Professor C. H. L. Goodman, Dr. F. C. Chen and Dr. P. W. McMillan for valuable discussions, Mr. R. Boone who helped to make some of the samples and take some measurements and Mr. R. Buxton of Loughborough University for his assistance in separating the fine copper and glass powders.

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Chapter 2

Carbon-Fibre Reinforced Composites with Zero Expansion

in the Low Temperature Range

by M. de F. F. Pinheiro and H. M. Rosenberg

2.1 Introduction

This chapter describes the thermal expansion behaviour of composites containing carbon fibre in an epoxy matrix in the range from above room temperature down to 20 K. Previous workers (Pirgon, Wostenholm and Yates, 1973) have shown that the thermal expansion of carbon fibre composites is quite small, but in one type of specimen we have found no detectable dimensional change from 20 up to 250 K.

Carbon fibre is composed of assemblies of graphite crystals which are aligned with their basal planes parallel to the length of the fibres. It is well established that in the temperature range up to about 400°C graphite has a negative coefficient of expansion within the basal planes although perpendicular to those planes the expansion is positive (Nelson and Riley, 1945; Sutton and Howard, 1962; Kellett and Richards, 1964; Entwisle, 1962). When graphite fibres are incorporated in a matrix with a positive expansion coefficient there will be a tendency for the dimensional changes of the fibres and of the matrix to cancel one another so that in one direction at least, a material of high dimensional stability is obtained (see Pirgon et al, 1973, for results down to 80 K).

2.2 Specimens and measurements

The specimens were made of Courtaulds Grafil carbon fibre incorporated in a matrix of epoxy resin - Shell Epikote 828 with NMA hardener. Two of the main types of carbon fibre available commercially were used. These have undergone differing heat treatments during manufacture which is optimised to produce a high tensile strength material (HT type) or a general purpose fibre (A type). The total volume concentration of fibre was in the range 58 to 73%. Four types of specimen were investigated. Two (one of HT and the other of A type) had the fibres laid in one direction only and the other two had the fibres arranged in thin layers with the orientation of the fibres in adjacent layers at 90° to each other. For the unidirectional specimens measurements of the thermal expansion were taken in directions both parallel to and perpendicular to the fibre axis. For the crossed fibre specimens measurements were only taken along one fibre direction. The measurements were made by incorporating the specimens into a capacity cell. The change in capacity, due to the change in length of the specimen was measured with a capacity bridge (White, 1961). In such an arrangement absolute changes in length of 0.1 nm or less can be detected quite easily. Several runs were taken on each specimen.

2.3 Results

Since the main interest, if these materials are to be used as constructional elements, is in the overall dimensional change when the specimen is cooled or heated, we present our results as the fractional change in length at a temperature \underline{T} , compared with the room temperature (293 K) length.

Figure 10 shows the results for both A and HT type specimens in the direction perpendicular to the fibre axis. The expansion is quite normal in both specimens. It flattens off below about 50 K and the overall dimensional change is slightly greater for the A type than for the HT type.

Figure 11 shows the results for HT type along the fibres both for the unidirectional (solid points) and for the crossed fibre (crosses) specimens. It will be noted that there is a net expansion when both these specimens are cooled from 293 down to 20 K although there is a minimum in the length at about 250 K. The expansion within the accuracy of our measurements is the same for both samples.

The results for the A type specimens shown in figure 12 are the most interesting. The unidirectional fibre specimens show a slight expansion on cooling down to 20 K with a minimum at 250 K. This is similar to the HT specimens. However, the crossed fibre specimens, after a small initial contraction, have no detectable dimensional change from 250 K right down to 20 K. The increased contraction in the perpendicular direction (figure 10) of the A type just cancels out the expansion along the fibres. The effective linear expansion coefficient over this range must be less than 5×10^{-8} K⁻¹.

2.4 Discussion

The main purpose of this paper is to present the results of some of our investigations and we defer a theoretical analysis to a later publication. The results shown in figure 12 are not necessarily the best in dimensional stability that can be achieved although they do seem to be better than those reported by other workers (Rogers et al, 1977). In particular the initial drop in the range from room temperature down to 250 K is a little disappointing and further investigations would be necessary to see if it can be suppressed by specimens of different composition and/or of fibre orientation. It should also be noted that above room temperature the crossed fibre specimens expand more than those with unidirectional fibres. This is because the expansion of those fibres which are at right angles to the direction of measurement is beginning to dominate. Nevertheless below room temperature the extremely high dimensional stability of the crossed fibre A type composites coupled with their lightness and their remarkable mechanical properties would suggest that they could be a very useful constructional material in cryogenic equipment.

2.5 Acknowledgements

We are most grateful to Dr. G. Gould and Dr. I. Thompson of Courtaulds Limited who prepared the samples for us. This work was done during the tenure of a scholarship by M. de F. F. P. from the Instituto Nacional de Investigação Científica, Portugal. A Preliminary Study of the Thermal Conductivity and

Expansivity of a Carbon-fibre/Magnesium-matrix Composite

by M. de F. F. Pinheiro, D. J. Radcliffe and H. M. Rosenberg

3.1 Introduction

An investigation has been made of one sample of a composite made from Thornel 50 carbon fibre in a magnesium metal matrix. The composite was made by melting the magnesium around the fibres with some Ti powder present. The fractional volume concentration of fibre was 0.35.

3.2 The experiments

Measurements were made of the thermal expansion of the composites in the direction parallel to the fibres in the temperature range 20-450 K. A standard capacitative technique was used to determine the change in length of the specimen as the temperature was varied.

The thermal conductivity was measured in the dimension parallel to the fibre axis from 3 K up to room temperature. A conventional Searle's bar technique was used and the temperature gradient along the sample due to a continuous heat input at one end was measured with Au-Fe/chromel thermocouples.

3.3 Thermal conductivity results

The thermal conductivity results are shown in figure 13. At low temperatures the conductivity is proportional to the absolute temperature. It passes through a maximum at about 25 K and it then decreases, passing through a shallow minimum at about 100 K. Beyond 100 K there is a slight increase of conductivity with temperature.

This behaviour is broadly that which one would expect for the conductivity of magnesium clone. This is not surprising since over most of the temperature range the conductivity of the carbon fibre is known to be very low (see our 1977 report). The only difference between the conductivity of the composite and that expected for a pure metal is the slight increase in conductivity with temperature beyond 100 K. In a pure metal the conductivity in this temperature region should be approximately constant. The rise in the conductivity of the composite at these temperatures would therefore appear to be due to the small contribution which is provided by the carbon fibres, because at higher temperatures this can become appreciable. Nevertheless this contribution is still quite small in absolute terms and the thermal conductivity of the composite is approximately equal to that of magnesium metal if one takes account of the fact that the effective cross-section of the metal itself is only about 65% of the geometrical cross-section of the specimen. Using this value for the cross-section, the calculated value of the conductivity using published data for the conductivity of magnesium, should be about 103 W m⁻¹ K⁻¹. This is to be compared with the value which we have measured for the composite at room temperature - 115 W m⁻¹ K⁻¹. This slightly higher figure is almost certainly due to the contribution of the carbon fibre.

Thus the conductivity of this composite at room temperature and above is reasonably good, it is about one quarter that of copper. It will, of course, have an electrical conductivity that is very similar to that of magnesium provided that the effective cross-section of the material is used as indicated above.

3.4 Thermal expansion results

The thermal expansion results are shown in figure 14 in which the relative change in length $\Delta \ell/\ell$ compared with the length at 290 K is plotted as a function of the temperature. The main point to note is that the overall expansion is very small. Between 290 K and 450 K the total dimensional change is 4 x 10⁻⁴. This yields an average expansion coefficient over this range of 2.5 x 10⁻⁶ K⁻¹. This is about 10 times less than that of pure magnesium (and about 7 times less than that of copper). The reason for this very low expansion is that up to about 400°C the thermal expansion of carbon fibre along the fibre direction is negative. This negative expansion therefore tends to counteract the positive expansion of the magnesium matrix and because the carbon fibre has such a high elastic modulus, the effect is to reduce the thermal expansion very considerably. A similar effect with carbon fibre composites which have an epoxy-resin matrix was described in our 1977 report.

In the low temperature region from room temperature down to 20 K the dimensional stability of the material is quite outstanding. Below about 250 K, the $\Delta l/l$ curve is more or less horizontal, indicating no measurable dimensional change (to within a few parts in 10⁵). The slight variations in the curve are outside our experimental error and are probably genuine effects. These are probably due to the fact that the contraction of the metal matrix and the expansion of the carbon fibre do not exactly cancel out at all temperatures. Nevertheless this flat curve at very low temperatures is very remarkable and although it is not quite so good as that described in Chapter 2 for crossed-ply carbon-fibre composites, it is still worthy of note. The magnesium composite however does have desirable characteristics which are not possessed by the epoxy-resin matrix composites. As already described the thermal and electrical conductivities are quite high - especially if they are compared with the values for carbon fibre in an epoxy matrix (see our 1977 report).

3.5 Conclusions

This preliminary investigation of the thermal properties of a magnesium/carbon-fibre composite shows that it is a potentially very useful lightweight material. It can presumably be used to much higher temperatures than can a conventional carbon-fibre/epoxyresin composite, it has very high dimensional stability and high electrical and thermal conductivities. Further investigations to study the mechanical properties, the effect of different fibre loading and also different types of fibre would certainly be worth consideration.

3.6 Acknowledgements

We are most grateful to Dr. A. Levitt for supplying the sample, to Dr. R. J. Weiss for helpful discussions and to Mr. R. Boone who took some of the measurements. M. de F. F. P. was in receipt of a grant from the Instituto Nacional de Investigação Cientifica, Portugal.

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Captions to Figures

- Figure 1 Thermal expansion of epoxy filled with copper spheres $(V_f = 0.30)$ vs. temperature. First heating.
- $\frac{\text{Figure 2}}{(V_{f} = 0.50)} \text{ thermal expansion of epoxy filled with copper spheres}$
- Figure 3 Thermal expansion behaviour of epoxy filled with copper spheres ($V_f = 0.30$) (125-150 µm). 1st to 4th heatings.
- Figure 4 Thermal expansion behaviour of epoxy filled with copper spheres ($V_f = 0.30$) (< 30 µm). Ist to 4th heatings.
- Figure 5 Thermal expansion behaviour of epoxy filled with copper spheres ($V_f = 0.30$) (> 30 µm). Slow and fast heatings.
- Figure 6 Thermal expansion of epoxy filled with copper spheres vs. temperature, below 293 K.
- Figure 7 Thermal expansion of epoxy filled with ballotini spheres $(V_f = 0.30)$ vs. temperature. Second heating.
- Figure 8 Thermal expansion of epoxy filled with copper spheres $(V_f = 0.30)$ vs. temperature. Comparison between theory and experiment.
- Figure 9 Thermal expansion of epoxy filled with ballotini spheres $(V_f = 0.30)$ vs. temperature. Comparison between theory and experiment.
- Figure 10 The fractional change in length, compared with the room temperature (293 K) length for A and HT type unidirectional carbon fibre composites in the direction perpendicular to the fibre axis. The volume concentration of fibres was 58% in both specimens.
- Figure 11 The fractional change in length, compared with the room temperature (293 K) length for HT type carbon fibre composite specimens. Circles, unidirectional fibres parallel to the fibre axis (volume concentration 65%). Crosses, 90° crossed fibres parallel to one set of fibres (volume concentration 73%).
- Figure 12 The fractional change in length, compared with the room temperature (293 K) length for A type carbon fibre composite specimens. Circles, unidirectional fibres parallel to the fibre axis (volume concentration 58%). Crosses, 90° crossed fibres parallel to one set of fibres (volume concentration 69.5%).
- Figure 13 The thermal conductivity of magnesium + 35% carbon-fibre composite from 3 to 300 K.

Figure 14 The thermal expansion of magnesium + 35% carbon-fibre composite from 20 to 460 K.



FIG. / Thermal expansion of epoxy filled with copper spheres ($V_f = 0.30$) vs. temperature. First heating.



 $(V_{f} = 0.50)$ vs. temperature. First heating.







FIG.4 Thermal expansion behaviour of epoxy filled with copper spheres ($V_f = 0.30$).



FIG. 5 Thermal expansion behaviour of epoxy filled with copper spheres ($V_{f}=0.30$).





FIG. 7 . Thermal expansion of epoxy filled with ballotini spheres (V_{g} = 0.30) vs. temperature. Second heating.





FIG. 9

Thermal expansion of epoxy filled with ballotini spheres ($V_{f} = 0.30$) vs. temperature. Comparison between theory and experiment.



Fig. 10



Fig.II





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