

Thermal Property Measurements and Data on Composite Materials

Summary

This Measurement Note reviews the experimental methods of obtaining thermal expansion and thermal diffusivity data on composite materials evaluated in a research programme at NPL under the title "Design with Composite Components", and gives example data for the various materials tested, including those from an interlaboratory exercise.

Thermal expansion measurements can be made by mechanical dilatometry, optical interferometry, or strain gauging methods. Each method has its strengths and limitations in terms of quality of data, repeatability and calibration issues. Factors affecting the results are internal stress distributions in the composites themselves, applied forces, and the representative nature of the volume of material being sampled by the technique.

Thermal diffusivity measurement is most conveniently made by the laser flash method, but this has limitations with composite materials possessing low values of thermal diffusivity because test-pieces may have to be of dimensions which are unrepresentative of the composite as a whole.

In each case the reliability of the data may be less than the stated potential error as a result of a combination of material and sample size effects, as well as issues of calibration.

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August 1997

Thermal Expansion

Definition

Linear thermal expansion is the fractional increase in length of a body when heated or cooled through a given temperature range, and is often quoted as a coefficient per unit temperature interval, either as an average over a stated range, or as the tangent to the expansion curve (*expansivity*) at a given temperature. The units are usually 10^{-6} K^{-1} .

Measurement techniques

Mechanical dilatometry

This is a well established technique, and commercial equipment is readily available. A test-piece in bar form is held against a stop in a support tube, and its free end is contacted by a push-rod. As the test-piece is heated and cooled its changes in length are transmitted by the push-rod to a measurement device, usually a displacement transducer.

Calibration routines are also well established, and have been incorporated into standards such as EN 821-1 for ceramic materials (but more widely applicable). The key factors in calibration are the absolute sensitivity of the displacement measuring device, the temperature homogeneity of the system, which can lead to a "baseline" shift (positive or negative apparent displacements with no test-piece present), and the correction for the expansion of the apparatus itself, that part against which the test-piece length is compared. It is necessary to set up a calibration procedure to identify these factors, to ensure that they are consistent, and to use them to correct the

direct output for an unknown to the true thermal expansion behaviour. There are several options for doing this. One possibility is to run the apparatus with two very different reference materials for which the expansion data are known. The difference in output is then directly related to the difference in responses of the apparatus for the two materials. The actual output of either reference material, but usually the one with characteristics closest to those of the apparatus material itself, can be used to obtain the apparatus expansion characteristic. Another is to calibrate the sensitivity of the measuring device directly using a micrometer to displace it known amounts, to run the apparatus with no test-piece to obtain the baseline shift, and to use a single reference material to determine the correction for the apparatus expansion. In either case, the reproducibility and reliability of the calibrations have limitations imposed primarily by the repeatability of mechanical contacts between the test-piece or reference pieces and the apparatus, which in turn relates to how the test-piece is held in the apparatus, and the conditions of its end contacts. These limitations are generally poorly understood, and lead to the quotation of results to unwarranted numbers of significant figures.

Typically, a mechanical dilatometer operated with care and attention to repeatability of calibrations and measurement procedures can achieve a reproducibility of about $\pm 0.2 \times 10^{-6} \text{ K}^{-1}$ measured over a range of 100 K, with an absolute accuracy of about $\pm 0.2 \times 10^{-6} \text{ K}^{-1}$. Thus over large temperature ranges, the absolute accuracy can be better than this, and over smaller temperature ranges, rather worse. The accuracy is best treated in this manner, because the limiting mechanical instabilities are present whatever the expansion coefficient of the material, and the sensitivity factor (a single temperature independent figure) can usually be determined rather more accurately than the other temperature dependent factors.

Interferometry

This is a potentially much more accurate method for determining thermal expansion. A coherent light source is split and reflected off surfaces at each end of the test-piece, and then recombined. As the test-piece expands, the level of interference between the recombined beams changes, and the total expansion can be determined by counting the number of dark fringes detected. One fringe corresponds to a test-piece end-face relative displacement of $\lambda/4$. For a calibrated wavelength, the method is potentially absolute, and needs no correction factors. With certain types of interferometer, e.g. the Jamin type which has been the subject of developments at NPL, displacements as small as $\lambda/1000$ can be resolved. However, the quality of the test-piece surfaces is a key issue. They normally need to be optically flat and reflective to act as their own mirrors. If this is impossible, as is usually the case with composite materials, mirrors need to be placed on the end surfaces, which poses questions of mechanical stability and correction for the expansion of the mirrors.

Other factors requiring consideration are alignment of the mirrors, temperature homogeneity of the test-piece and the temperature stability needed to obtain stable fringes. In addition, the measurement normally has to be conducted in vacuum to avoid the distorting effects of convection in air along the beam path. The technique is best suited to accurate measurements near room temperature, particularly on low thermal expansion materials, but is routinely used at NPL for calibration of expansion coefficients of steel gauge blocks [1].

Strain gauges

This relatively little-used technique [2] has been evaluated at NPL on composite materials and found to be quite promising provided care is taken over calibration routines [3]. The technique used employs a thermal expansion reference material with an identical strain gauge attached to it and wired so that the differential expansion is recorded (x,y). There are considerable advantages in this technique for examining the spatial homogeneity of expansion or dimensional stability of large pieces inappropriate for dilatometry or interferometry. The main drawback is the temperature limitation of the gauge and the means of securing it to the material. Conventional foil gauges can be used typically to 200°C, and although higher-temperature versions and appropriate adhesives can be obtained, they are relatively expensive and specialised, so that 200°C should be taken as the upper temperature limit.

It is essential that identical installation techniques are employed on the test-piece and on the reference, and that the reference is well characterised over the required temperature range. In the tests they must also be at identical temperatures, and thus this technique should ideally be performed at a series of temperature holds with stabilisation periods, unless the test-pieces have sufficiently small thermal masses that the test can be done reliably whilst temperature ramping.

The technique has been validated at NPL using copper and aluminium with ULE silica (Corning Glass Works, near zero expansion coefficient glass) as the zero reference [4]. Questions of the lateral sensitivity of strain gauges do not apply to isotropic materials as the test-pieces are not mechanically strained, and any secondary effects are eliminated in the difference calculations. Comparisons between mechanical dilatometry and the strain gauge technique have been carried out on MMCs, and a good match was found to 200°C, above which the properties of the gauge and/or its adhesive affected the results. With anisotropic materials, lateral sensitivity is more important

[2], and increases the potential errors in the results.

Measuring thermal expansion properties of composites

General

One of the key points arising from an interlaboratory study [5] is the importance of the composite test-piece having a representative volume. Interruption of the reinforcement by cutting free surfaces influences the internal stresses, and this can influence the thermal expansion recorded. The test-piece therefore needs to be large enough that the reinforcement can be considered substantially homogeneous and that effects of the cut edges and the local inhomogeneities can be ignored. Preferred practice at NPL for in-plane measurements on continuous fibre PMCs is to use a square plate test-piece. The effect is not thought to be significant in homogeneous particulate reinforced materials. Further, in long-fibre multi-ply materials it seems to be important to ensure that the through-thickness structure is symmetrical, so that no bending occurs. Normally, composites are designed with symmetrical lay-ups to prevent this, but manufacturing processes do not always guarantee perfect balance. Machining test-pieces to a less than normal thickness will unbalance the structure, and could lead to bending during thermal cycling.

Mechanical dilatometry

The push-rod in a mechanical dilatometer normally applies a compressive axial force on the test-piece to maintain constant contact. Typically this force is 0.5 – 2 N, but may be less if a balanced thermomechanical analyser type of apparatus is used. This force may be sufficient to induce permanent distortion in composites when the matrix phase softens. In polymer-based systems, this can be upwards of 150°C, and in aluminium alloy based metal-matrix composites, above about 450°C. The distortion may be limited to "bedding down" of the contact surfaces, but in some cases, may induce bulk creep effects. The progressive "ratchetting" seen in repeat thermal cycling of some composites (e.g. [Figure 1](#)) may in part be attributable to creep effects. The effects tend to be negligible parallel to the reinforcement directions in long-fibre composites, but may be considerable across ply structures in 2D laminates.

Interferometry

Separate mirrors have to be used because composites cannot be adequately polished to give reflective surfaces. Resting the test-piece stably on a mirror and placing a second mirror on the top surface is the simplest configuration to use. The test-piece surfaces need to be accurately flat and parallel, and as-moulded surfaces may have to be machined to achieve the alignment conditions required for the mirrors. Care is needed not to unbalance systems in such a machining process. Typically, test-pieces might be quite small, but for larger items made from low thermal expansion materials, carefully aligned mirrors can be glued to a face of the composite in order to achieve representative lengths over which to measure expansion.

Strain gauges

The area covered by a strain gauge is usually quite small, 1-5 mm, and this means that it samples the local behaviour rather than the longer range average. In composites which have homogeneous reinforcement, such as flat laminates, unidirectional reinforcements and particulate composites, the technique can be used reliably (see [Figure 2](#)), but caution needs to be exercised with woven structures. The active part of the strain gauge needs to cover several representative reinforcement units for the results to be representative of bulk material behaviour.

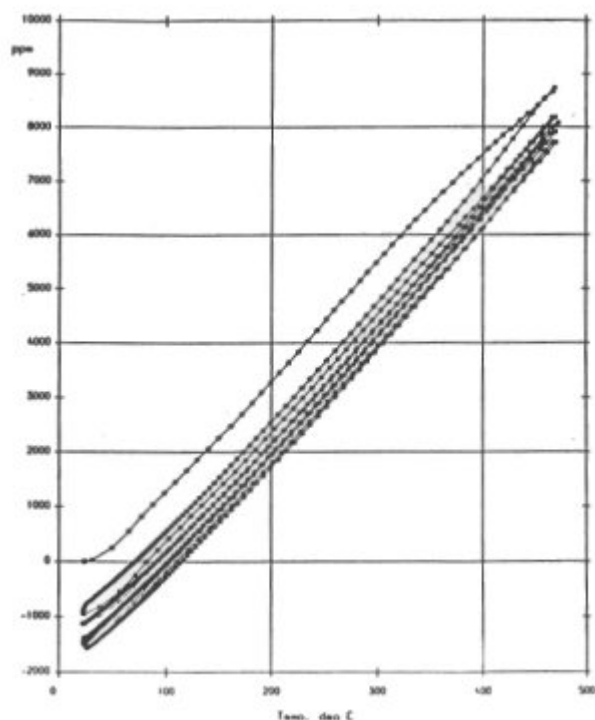


Figure 1 - Thermal expansion behaviour on cycling of an MMC determined by mechanical dilatometry.

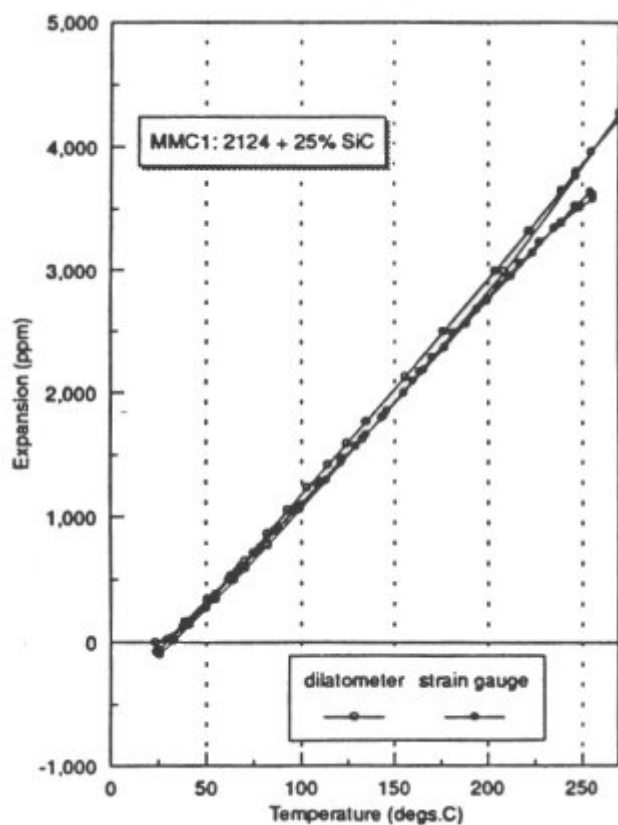


Figure 2 - A comparison between strain gauging and mechanical dilatometry applied to thermal expansion measurement of an MMC.

Table 1 - Typical thermal properties of some representative composites determined in NPL's programme

Material type		Thermal expansion coefficient, 10^{-6} K^{-1} over temperature range			Thermal diffusivity, $10^{-6} \text{ m}^2 \text{ s}^{-1}$, isotropic or through- thickness
		Isotropic	Parallel to reinforcement	Perpendicular to reinforcement	
MMCs	2124 Al alloy + 25% SiC, as manufactured (extruded)	16.5 ± 0.3 (25 - 200°C)	-	-	60 ± 2 (25°C) 50 ± 2 (400 ° C)
	6061 Al alloy + 20% Al_2O_3 solution treated and aged condition T5	19.5 ± 0.5 (25 - 200°C)	-	-	61 ± 2 (25°C) 50 ± 4 (400 ° C)
	2124 Al alloy, 17% SiC, rolled strip, as manufactured	17.1 ± 0.2 (25 - 200°C)	-	-	-
	Ti alloy/Sigma SiC fibres, unidirectional sheet	-	6.8 ± 0.2 (25 - 200°C) 7.2 ± 0.2 (25 - 400°C)	8.0 ± 0.2 (25 - 200°C) 8.6 ± 0.2 (25 - 400°C)	-
	Ti alloy/Sigma SiC fibres, crossply sheet	-	6.8 ± 0.2 (25 - 200°C) 7.4 ± 0.2 (25 - 400°C)	-	-
PMCs	Woven 0-90° crossply carbon/epoxy mould material, 2/2 twill carbon fabric with 1-8-1 lay-up, outer plies with a finer weave than inner plies, thickness 6 mm with as-moulded surfaces	-	2.6 ± 0.3 (25 - 100°C)	50.0 ± 0.5 (-25 - +25°C) 57.5 ± 2.0 (25 - 100°C)	0.38 ± 0.02 (25°C) 0.30 ± 0.02 (200°C)
	Woven 0-90° crossply glass/epoxy mould material, glass fabric same lay-up as LTM10/carbon above	-	10.0 ± 0.5 (25 - 100°C)	36.0 ± 0.5 (-25 - +25°C) 50.0 ± 1.0 (25 - 100°C)	0.38 ± 0.02 (25°C) 0.30 ± 0.02 (200°C)
CMCs	Tyranno SiC fibre, 50 v/o, BMAS glass-ceramic matrix, 0 - 90° balanced cross-ply	-	3.5 ± 0.1 (25 - 1000°C)	-	0.72 ± 0.02 (100°C) 0.65 ± 0.02 (1000°C)
	Carbon/carbon balanced cross-ply (typical)	-	0.4 ± 0.2 (25 - 500°C) 1.0 ± 0.2 (25 - 1000°C)	13.0 ± 0.5 (25 - 500°C) 13.5 ± 0.5 (25 - 1000°C)	-

Composite characteristics

Basic average thermal expansion data for a range of composites is shown in [Table 1](#). However, presenting the data in this form disguises a number of features which may occur:

- **Annealing.** The composite test-piece, as prepared, has been cooled from its manufacturing temperature at an uncontrolled rate which is different from the conditions then being used for thermal expansion measurement. During the first thermal cycle permanent changes can occur which can be attributed to annealing of residual stresses on heating and regenerating them at different levels during controlled cooling. It is usually necessary to ignore the first thermal cycle while the test-piece becomes normalised to the testing rate.
- **Low temperatures.** With particular regard to PMCs, reducing the temperature substantially below room temperature may induce permanent changes to the composite behaviour as a result of an increase in matrix microcracking caused by increased thermal strain mismatch, hence changing the net thermal expansion behaviour over the entire temperature range.

- **Hysteresis.** The length achieved at a given temperature may depend on whether the sample was heated or cooled to that temperature. This arises due to the effects of the build-up or release of internal stresses having different functions of temperature. The result may be that a loop develops on cycling. Mostly, data are quoted as average behaviour, and the loop width is unaccounted for.
- **Ratchetting.** With cycling, there may be a progressive net increase or decrease in the length of the test-piece. This originates from permanent changes being accumulated in the structure of the composite, such as the development of delaminations at interfaces or accumulated thermal strain induced plastic deformation of matrices.
- **Ageing.** Prolonged heating or thermal cycling can produce changes to average expansion coefficients as a result of changes in interface characteristics. Progressive development of interface phases can occur. The effect is temperature dependent, but no changes in expansion characteristics were found following ageing of several aluminium alloy/SiC MMCs up to 5000 h at 250°C, which might be considered to be an upper use temperature for long-term mechanical performance. In PMCs progressive changes in the cross-linked structure of the matrix can alter thermal expansion coefficients and effective glass transition temperatures.

The magnitude of the above effects varies from material to material, and no single guideline is applicable. It is recommended that when thermal expansion data are critical to an application, any relevant effects are investigated systematically.

Thermal Diffusivity

Definition

Thermal diffusivity (a) is a measure of the rate at which heat can flow through a body whilst changing its temperature, and is related to the steady state thermal conductivity (λ) through the relation:

$$a = \lambda / \rho c_p$$

where ρ is the density and c_p is the specific heat. It has units of $\text{m}^2 \text{s}^{-1}$.

Measurement techniques

The pulse method is well established, has been extensively researched over many years, and has been widely applied to composites. A short-duration heat pulse is applied to one face of a thin parallel-faced disc test-piece, and the time profile of the response on the other face is monitored. The thermal diffusivity is calculated from the half-rise time of the transient. It is usually necessary to apply a black coating to both faces to maximise heat absorption and minimise direct transmission. The method has been standardised in EN 821-2, which provides an operating framework, although the calculation routines are not standardised, and leave many options open for individual methods of data analysis. Typically, corrections are made for finite pulse time and radiation losses. This type of technique often uses a laser pulse and an infrared sensor, but alternatively, a high-intensity flash lamp and a thermocouple can be used. The flash method has considerable time and cost advantages compared with traditional methods of thermal conductivity measurement which require an equilibrium thermal gradient to be achieved through a larger test-piece. The technique does not require calibration as such because the principal measurements are transient half-rise time and test-piece thickness. Checks can be made using test-pieces cut from thermal conductivity reference materials, but these do not cover a wide range of thermal diffusivity. Overall, confidence in the result of test is said by users to be typically $\pm 4\%$, although a recent interlaboratory study suggested that this may be optimistic [5].

Measuring thermal diffusivity of composites

This method is ideal for through-thickness measurement, but the required shape of test-piece usually rules out in-plane measurements in reinforced materials unless built-up specimens comprising several thicknesses are used. Alternatively, transient lateral heat flow methods are a possibility for very thin sheet, but are not well established.

A key factor in obtaining reliable measurements is the use of a representative test-piece. Since the illumination diameter in most systems is limited by the laser beam dimensions to about 12 mm, and the detection times require test-pieces of 0.5 to 3 mm thick, the reinforcement in the test-piece needs to be homogeneous and representative within this volume. For particulate reinforced MMCs this requirement poses no problems, but is significantly limiting

for long-fibre reinforced materials. For an in-plane reinforced test-piece measured through thickness, if an infrared detector is used which covers most of the rear face of the test-piece, this will average the response from different parts of the disc area, but if a thermocouple is used, this measures the temperature at a point, and could be strongly influenced by the local reinforcement. Thus for i.r detectors, the periodicity of the reinforcement needs to be typically less than 3 mm, but less than 1 mm for thermocouple detectors. The thickness also needs to contain representative reinforcement, preferably at least four plies. If the requirement for an acceptable transient time requires a test-piece which is thin, this condition may not be achievable.

In an interlaboratory study it was found that, using the laser flash/i.r. detector method, a CFRP could not reliably be measured through-thickness at 3 mm thickness (half the composite panel thickness), and measurement was impossible in a similar GRP. They needed a xenon flash lamp/thermocouple technique. The laser flash method needed a test-piece thickness of < 1 mm to achieve an appropriate level and time period of detection without damaging the test-piece surface, which would not be representative of the ply construction [5].

Thus, the reliability of measurements depends very much on the material type and on whether an appropriate test-piece suitable for both the available system and the reinforcement structure can be prepared.

Composite characteristics

Table 1 includes some typical data determined for materials in the recent NPL Design of Composite Components (DCC2) programme, with error bars based on identified local variations and potential errors of measurement. Thermal diffusivity usually declines somewhat with increasing temperature over the normal use range. Some major points not covered by these data are:

- As with thermal expansion, there can be annealing, hysteresis and ageing effects during thermal cycling which are detectable by thermal diffusivity measurement [6] provided that care is taken concerning reliability of measurement and stability of temperature before measurement. Hysteresis tends to be less than 2% of the measured value.
- Unless a thick cross-section is available, measurement by laser flash parallel to the reinforcement requires a built-up test-piece subsequently sectioned to produce a thin disc. Clamping may be an inadequate technique, and adhesives have to be selected with care, especially for high-temperature measurement.
- With open porous materials, or ones in which debonding has occurred along fibres, the test atmosphere may have an influence. To obtain consistent data may require the test to be performed in vacuum.

Acknowledgements

This work forms part of the Design with Composite Components Programme 1993-7 supported by the Department of Trade and Industry under the Materials Metrology Programme. In addition to NPL, contributions to the work have also been made by AEA Technology Ltd and UMIST Materials Science Centre.

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CMMT(MN)011

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August 1997

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