THERMAL DIFFUSIVITY MEASUREMENTES ON FIBRE REINFORCED POLYMER THROUGH THE STEP CHANGE TECHNIQUE.

P. Di Modica[a]*, P. Vollaro[b], A. G. Gibson[a]

[a]Centre for Composite Materials Engineering, School of Mechanical & Systems Engineering, Newcastle University, Stephenson Building, Claremont Rd., Newcastle upon Tyne, NE1 7RU UK.
[b] 1Technological District on Engineering of Polymeric and Composite Materials and Structures (IMAST), P.zzle Enrico Fermi 1, 80055 Portici, Naples, Italy.
**e-mail address of the corresponding author p.di-modica@ncl.ac.uk, pietrodimodica@hotmail.it*

Keywords: CFRP, Composite, Thermal diffusivity, GFRP

Abstract

Thermal diffusivity of CFRP and GFRP, courtesy of Cytec, has been measured through a step change technique developed at Newcastle University. The step change technique is capable of measuring thermal diffusivity without the need of knowing or estimating the heat capacity (density and specific heat). Results from room temperature to 100°C for unidirectional CFRP and 0/90 GFRP supplied by Cytec are presented. A comparison between the measurements obtained through the step change technique and the guarded heat flow meter method, is presented and good agreements were found.

Introduction

Heat transport properties are very important for understanding composites behaviour in fire. Composites thermal conduction is quite low compared to metals but unfortunately composite materials degrade and lose strength at temperature much lower than metals. Thermal diffusivity is the ratio of thermal conductivity, specific heat and density. Knowing thermal diffusivity as function of temperature is very important because it is the heat transport property used in FEM codes like ANSYS for thermal calculations [1]. Thermal diffusivity can be measured using the TPS method, which we are not going to deal with, or thorough the measurements of all the three properties previously mentioned. The aim of this paper is to show a technique that can estimate thermal diffusivity of composites or any material without the need of measuring any of the three thermal and physical properties previously mentioned.

Step-change method

The step change method developed in Newcastle University is based on the solution of the Laplace's equation, in the case of 1-dimensional heat flow in the laminate z-direction (through thickness direction), equation (1).

$$\dot{T} = \frac{1}{\rho(T)C_P(T)} k_z \frac{\partial^2 T}{\partial z^2} = \alpha_z \frac{\partial^2 T}{\partial z^2} \tag{1}$$

Where ρ is the density [kg m^{-3}], C_P is the specific heat [J kg^{-1} K^{-1}], k_z is the thermal conductivity in the generic z-direction [W m^{-1} K^{-1}], α_z is the thermal diffusivity in the generic z-direction.
The solution of a step change experiment can be easily expressed in terms of the dimensionless centre line temperature, given by:

$$\theta(t) = \frac{T_\infty - T(t)}{T_\infty - T_0} \tag{2}$$

where $T(t)$ is the centreline temperature at time t, T_0 is the initial uniform temperature in the slab and T_∞ is the temperature suddenly imposed at the slab surface. θ, therefore, varies from 1, at the start of the test, to 0, at long times, regardless of whether the slab is heated or cooled. The principal factor determining the variation of temperature with time is the Fourier number, which is given by:

$$Fo = \frac{\alpha t}{b^2} \tag{3}$$

b is the slab half-thickness [m] (the distance from the surface to the centreline), t [s] is time and α is the thermal diffusivity [m^2 s^{-1}] in the through-thickness direction. The Fourier number can be regarded as a dimensionless measure of time.
In the well-known solution of equation (1), for the case of the centreline temperature of an infinite plate, the centreline temperature of the slab is given by the Fourier series:

$$\begin{aligned} \theta = \frac{T_\infty - T}{T_\infty - T_0} &= \frac{4}{\pi}\exp\left(-\frac{\pi^2 Fo}{4}\right) - \frac{4}{3\pi}\exp\left(-\frac{9\pi^2 Fo}{4}\right) \\ &\quad + \frac{4}{5\pi}\exp\left(-\frac{25\pi^2 Fo}{4}\right) - \frac{4}{7\pi}\exp\left(-\frac{49\pi^2 Fo}{4}\right) \ldots\ldots \end{aligned} \tag{4}$$

For values of Fo exceeding ~0.2 this series can be truncated to the first term without significant loss of accuracy, so:

$$\theta = \frac{4}{\pi}\exp\left(-\frac{\pi^2 Fo}{4}\right) \tag{5}$$

This solution is restricted to the situation where the surface temperature is brought instantaneously to T_∞. In practice it is seldom possible to change the surface temperature quickly enough to realise this boundary condition exactly. When the temperature change is accomplished through contact with a fluid, the rate of heat flow into or out of the solid is influenced by the coefficient for convective heat transfer at the surface. The relative importance of the resistance to surface heat transfer and the resistance to heat flow through the solid are described by the Biot number, which is given by:

$$Bi = \frac{hb}{k} = \frac{hb}{\rho C_P \alpha} \tag{6}$$

where h is the surface convective heat transfer coefficient [W m^{-2} K^{-1}] and k is the thermal conductivity of the material at the surface [W m^{-1} K^{-1}]. Bi can be regarded as the *resistance to internal heat flow*, divided by the *resistance to external heat flow*. A large value of Bi, exceeding about 100, would be required for equation (5) to apply with sufficient accuracy to enable the thermal diffusivity to be calculated from such a step change experiment. A small value of *Bi*, less than about 0.1, would correspond to the case where surface heat transfer was the main limiting effect. In this case the main temperature change would be through the film of fluid at the surface, with very little temperature variation across the solid. Although efforts were made in the present work to maximise the surface heat transfer coefficient the situation of $Bi > 100$ is difficult to achieve, so it was necessary to allow for the effect of surface resistance to heat flow. Heisler [2] provided an analytical modification to equation (1) that allows for this. In the case where $Fo > 0.2$, equation (5) for the centreline temperature with Heisler [2] modification becomes:

$$\theta = C \exp\left(-\zeta^2 Fo\right) \tag{7}$$

where ζ and C are functions of Bi. ζ relates to the roots of the following equation:

$$Bi = \zeta \tan \zeta \tag{8}$$

and

$$C = \frac{4 \sin \zeta}{2\zeta + \sin 2\zeta} \tag{9}$$

Equation (7) implies a linear relationship between log(θ) and the Fourier number. It forms the basis of the well-known Heisler plots [2, 3], which were widely used for heat transfer calculations, and which still appear in many heat transfer books. Equation (8) poses the minor difficulty of having Bi as a function of ζ, rather than the reverse. The actual values of Bi encountered in the present study were in the range, 0.3-80. For $-3 < \ln\left(Bi\right) < 3$, which correspond to $0.05 < \ln\left(Bi\right) < 20$, Liukov [4] proposed the following expression as an approximation of the first root of equation (8):

$$\zeta = \frac{\pi}{2} \Big/ \sqrt{1 + \left(\frac{2.24}{Bi^{1.02}}\right)} \tag{10}$$

Although the equation is stable even for very small Bi, it gives acceptable values of ζ for $Bi \geq 100$. Below that the error in the estimate of ζ is greater than 1%. It is unknown the error introduced in the calculation of the Fourier coefficients, equation (9). Yovanovich [5] showed that equation (11), reported below, is capable of calculating with great accuracy the value of ζ for very big or very small Bi regardless of the values of the parameter n. In the intermediate range a suitable value of the parameter n has to be estimated to obtain an accurate relationship between ζ and Bi.

$$\zeta = \frac{\pi}{2} \Bigg/ \left[1 + \left(\frac{\pi}{2} \Big/ \sqrt{Bi} \right)^{n} \right]^{\frac{1}{n}} \tag{11}$$

In particular, in the range of $0.5 < Bi < 5$, ζ can be accurately described, with an error of less than 0.4%, by the following expression:

$$\zeta = \frac{\pi}{2} \Bigg/ \left[1 + \left(\frac{\pi}{2} \Big/ \sqrt{Bi} \right)^{2.139} \right]^{\frac{1}{2.139}} \tag{12}$$

Re-arranging equation (7) gives the following direct relationship, which can be used to determine α from a step-change experiment.

$$\alpha = -\frac{b^2}{\zeta^2 t} \ln\left(\frac{1-\theta}{C} \right) \tag{13}$$

corresponding to θ values of 0.4, 0.3 and 0.2. Equation (13) was used to find three values of α for each set of measurements, from which the mean value of α was taken. This relationship applies for heat flow in any of the three principal laminate directions, as long as one-dimensional heat flow conditions are achieved. The calculation process involves an iterative step, since, from equation (5), the thermal diffusivity or conductivity of the material must be known in order to calculate the Biot number. The iterative procedure is the following:

1. Guess/estimate of a value of ζ;
2. Calculate Bi and C using equation (8) and equation (9) respectively;
3. Calculate the three thermal diffusivities corresponding to the times of the 40% 30% and 20% of the dimensionless temperature curve using equation (13);
4. Calculate the averages of the three thermal diffusivities;
5. Calculate the thermal conductivity if the density and specific heat are known, equation (6);
6. Calculate Bi, equation (6);
7. Calculate ζ, equation (12);
8. If the initially estimated ζ is the same of the calculated one then the iteration is finished, otherwise restart from point 1 with a different value;

It has to be highlighted that known or estimated values of density and specific heat are required for this calculation at point 5. Considering the density as constant does not reflects on big errors. Knowing specific heat versus temperature is very important to have good

estimates of the thermal diffusivity. Another parameter that has to be known or measured for this technique to be used is the convective heat transfer coefficient at the point 6.
A flat slab of a material with a high conductivity, like an aluminium slab, can be used to measure the convective heat transfer coefficient provided that the step change is small enough to consider Newton's law of cooling applicable. The slab can be considered as a lumped parameter system. In this case the solution of the dimensionless temperature can be written in the following exponential form:

$$\theta = \exp\left(-\frac{h}{b\rho C_p}\right) \tag{14}$$

Rearranging equation (14) it can be shown that a linear relation exist between $\ln(\theta)$ and $-h/b\rho C_p$. With a simple linear regression the value of the convective heat transfer coefficient can be calculated [6]. To avoid measuring or estimate the density and specific heat another procedure could be applied. The use of 2 samples of the same material but of two different thicknesses would allow finding the thermal diffusivity ignoring the heat capacity (ρC_P) of the material. The steps are the following:

1. Guess/estimate Bi for the thinner (thicker) sample;
2. Calculate ζ and C through equation (12) and equation (9) respectively;
3. Calculate the three thermal diffusivities, equation (13), corresponding to the times of the 40% 30% and 20% of the dimensionless temperature curve;
4. Calculate the averages of the three thermal diffusivities;
5. Scale up Bi for the thicker (thinner) sample using equation (6);
6. Calculate ζ , C and α as in points 2 to 4 for the thicker (thinner) sample;
7. Compare the thermal diffusivities found for the 2 different thicknesses and if necessary restart from point 1 with a different value of Bi ;

The convergence of the thermal diffusivities is found using Newton-Raphson method. The drawback of this technique is that the starting time (step time application) of the transient response has to be accurately recorded for both experiments regarding the 2 different thicknesses otherwise the procedure will not converge with a good accuracy on the thermal diffusivity.

Materials and methods

Two different materials were used: a pre-preg unidirectional carbon-epoxy system, MTM44-1FR® courtesy of Cytec®, and a pre-preg 0/90 glass fabric-phenolic system, MTM82S® courtesy of Cytec®. Due to the orthotropy of the material, measurements in 2 directions for each material were required. The samples given by Cytec were in the shape of flat slab of 100mmX100mmX 10mm, see **Figure 1** a), so to measure the properties in the in-plane direction (along the fibres for what concerns the CFRP) the cut of the slab and reassembly of every stripe was necessary, see **Figure 1** b). The samples were instrumented with a thermocouple type K measuring the centre line temperature and insulated all around with 10mm thick CEMTHREM® board, see **Figure 1**. To favour the thermal contact between the thermocouple and the specimen a heat transfer compound was used.

To apply the step change the samples were left over night in temperature controlled chamber at a certain temperature (T_0) to reach thermal equilibrium and then very quickly transferred in a digital temperature controlled agitated water bath at the other temperature of the step (T_∞). The recirculation of the water insured a convective heat transfer coefficient high enough to apply the procedure. The measured property will be attributed to the average of the temperatures used for the step change. The step has to be small enough to consider the thermal diffusivity constant during the experiment and at the same time large enough to be sensed by the specimen and registered by the thermocouple. Acceptable step amplitude is between 10°C and 20°C. To record the transient thermal response an IOTech DAQ Shuttle 55 was used. During each experiment 2 channels were recorded at the same time at 4 Hz: the sample centre line temperature and the water bath temperature. The scan corresponding to being in touch with the water was noted on a log book. It represents the starting point of the transient response and so of the analysis of the results. Three replicates for each specimen at each step have been performed.

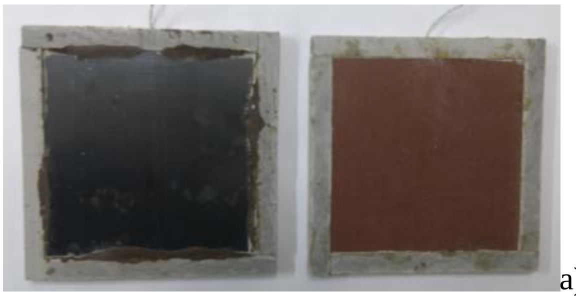
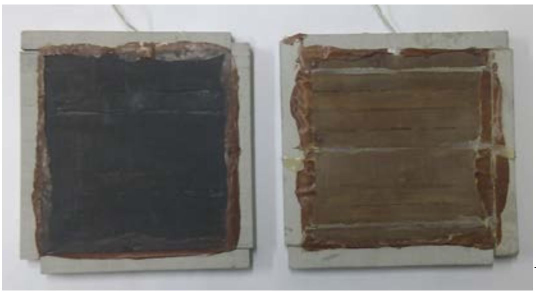

a)

b)

Figure 1: Samples for: a) the through-thickness direction measurements; b) in-plane direction measurements.

Results and discussions

As it can be seen in the following figures the thermal diffusivity tends to increase with increasing temperatures with the exception of the CFRP results, **Figure 2** a), in the through-thickness direction. This means that the higher is the temperature the less heat the CFRPs conduct in the through-thickness direction. The comparison between the two different ways of implementing the step change technique has been performed only with the CFRP in the through-thickness direction because the other samples were not ready at the time of the experiments. It can be observed that the thermal diffusivity of fibre reinforced polymers in the direction orthogonal to the fibres has comparable values or at least has the same significant figures among different fibre reinforced polymers. The exception is the thermal diffusivity of the CFRP sample in the direction along the carbon fibres because of their very high conductivity compared to glass fibres, an amorphous material.

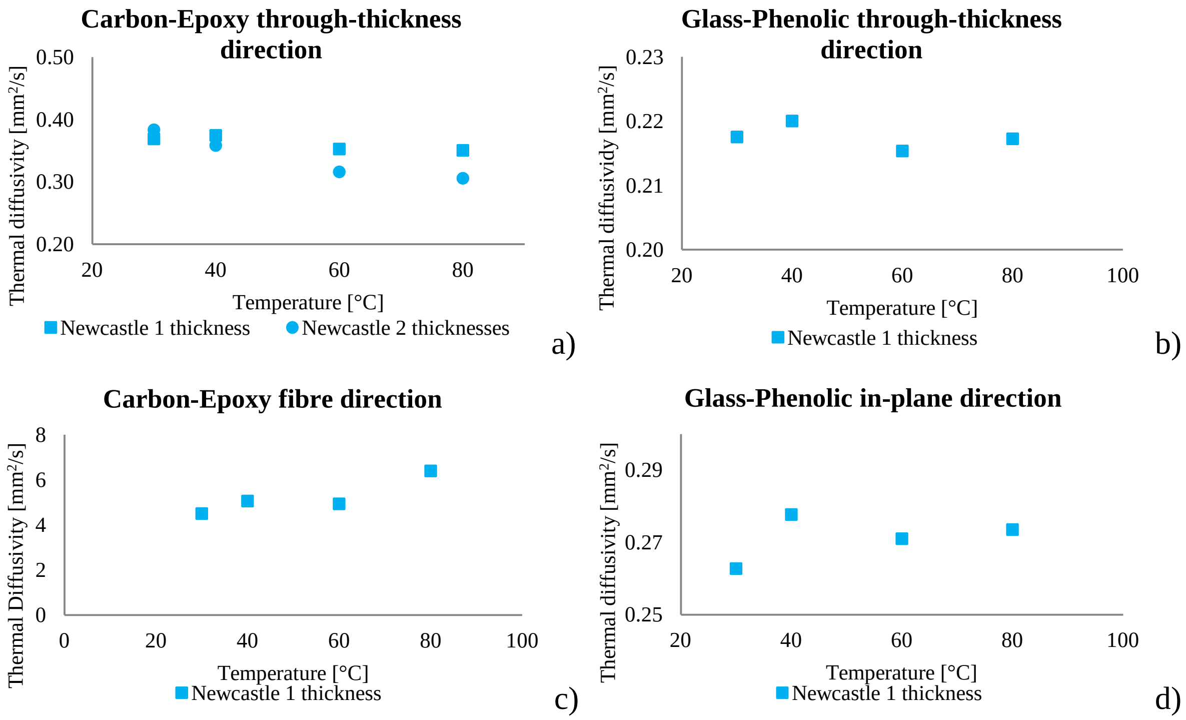


Figure 2: Thermal diffusivity of GFRP and CFRP in the in-plane through-thickness direction; a) reports the measurements with 2 different thicknesses.

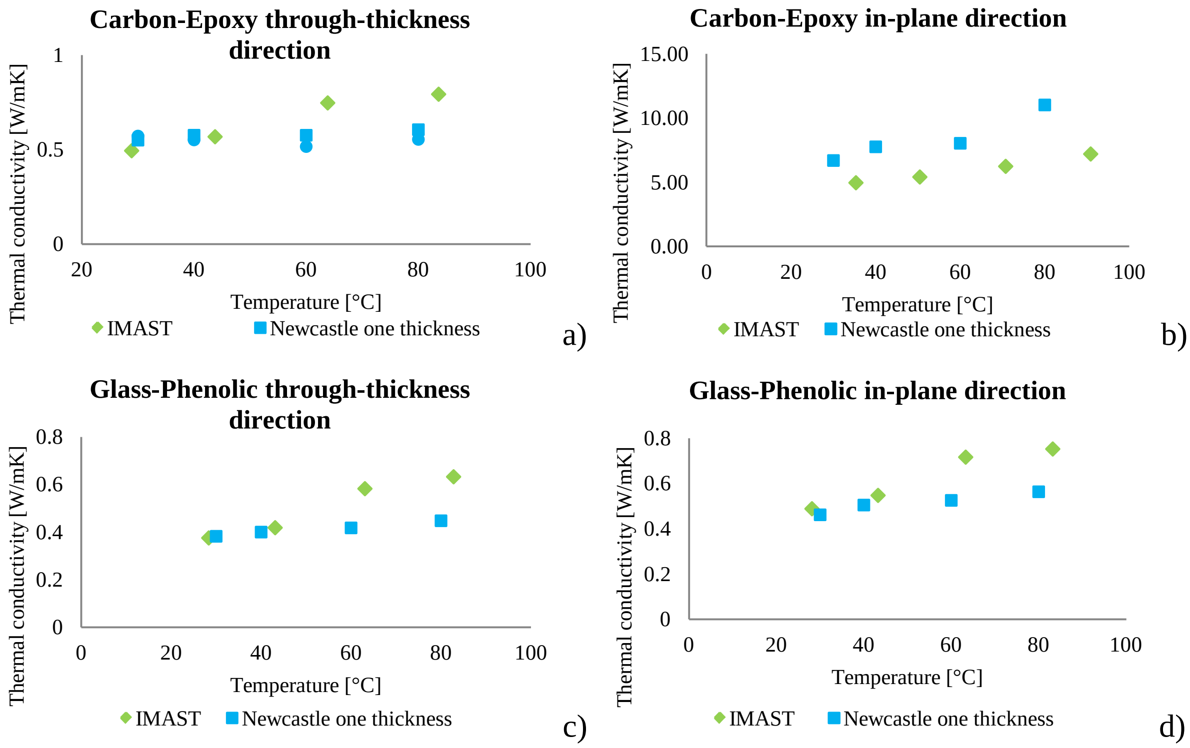


Figure 3: Thermal conductivity of GFRP and CFRP in the through-thickness and inplane direction.

The UNITHERM™ MODEL 2022 [7] is capable of measuring thermal conductivity at constant temperature [8]. In order to compare the 2 methods, density and C_p needed to be evaluated at different temperatures. Density has been considered constant over the range of temperatures used in this work. The following expressions for C_p VS temperature have been used:

$$C_p = 3.1 * T + 885 \tag{15}$$

$$C_p = 3.6 * T + 950 \tag{16}$$

for CFRP and GFRP samples respectively (source internal SP report of FIRE RESIST project). Good agreement is shown in **Figure 3** despite the different measuring methods in terms of physical principle and equipment accuracy.

Conclusion

Thermal diffusivity measurements have been performed using an innovative technique, the step-change method. Comparison with a standardised measuring method has been performed and the results showed good agreement. Thermal diffusivity tends to increase with increasing temperature with the exception of CFRP in the through-thickness direction. GFRP thermal diffusivity does not change much in different directions whereas CFRP thermal diffusivity is strongly anisotropic with the thermal diffusivity along the fibres direction being 10 to 12 time higher than the one across the fibres, as it can be noticed comparing **Figure 2** a) and c). The marked anisotropy of carbon fibres is the reason of this behaviour. Future development is needed to use this technique with lower Bi or changing convective heat transfer coefficient

Acknowledgements

The authors acknowledge FIRE-RESIST EU246037, a European FP7 project, for the thermal diffusivity and C_p measurements and the COCET - PON020000293206086 project for the thermal conductivity measurements.

References

1. Urso Miano, V., *Modelling composite fire behaviour using apparent thermal diffusivity*, in *University of Newcastle upon Tyne. School of Mechanical and Systems Engineering*
2011, University of Newcastle upon Tyne: Newcastle upon Tyne. p. ix, 114.
2. Heisler, M.P., *Temperature Charts for Induction Heating and Constant-Temperature*, in *Trans. ASME*. 1947. p. 227-236.
3. Cengel, Y.A., *Termodinamica e trasmissione del calore*. 1998: McGraw Hill.
4. Liukov, A.V., *Analytical Heat Diffusion Theory*. Academic Press, 1968.
5. Yovanovich, M.M., *Simple Explicit Expressions for Calculation of the Heisler-Grober Charts*. 1996 ASME National Heat Transfer Conference, Thermophysics and Thermophysical Properties Session, Houston, Texas, Aug. 3-6, 1996.
6. Cengel, Y.A. and M.A. Boles, *Thermodynamics : an engineering approach*. 3rd ed. McGraw-Hill series in mechanical engineering. 1998, Boston: WCB/McGraw-Hill. xxxix, 1010.
7. UNITHERM, *THERMAL CONDUCTIVITY INSTRUMENT*, in *User's manual*. 2006, ANTER CORPORATION, 1700 Universal Road, Pittsburgh, PA 15235-3998, (412) 795-6410.
8. ASTM E1530 – 11, A.I., *ASTM E1530 – 11, Standard test method for evaluating the resistance to thermal transmission of materials by the guarded heat flow meter technique*.