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A Method for Measuring the Thermal Diffusivity of Intermediate Thickness Surface Absorbing Samples and Obtaining the Ratio of Anisotropy by the Converging Wave Flash Method

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Abstract The converging thermal wave, flash technique for measuring thermal diffusivity is suitable for use on samples that are sufficiently thick or thin in comparison to the annular heat source, to be described by a three-dimensional or two-dimensional approximation of the heat conduction equation, and sufficiently absorbing to ensure generation of a heat source at the surface. However, samples of intermediate thickness, which lie between these regimes, cannot be analyzed. In this article, heat diffusion in the samples of varying thicknesses is modeled, and a semi-analytic expression is used to describe the dimensionality of any thickness, allowing the converging wave method to be extended to intermediate thickness samples. Applying the analysis to anisotropic samples, a method is proposed to find the anisotropy ratio of the in-plane to perpendicular-to-plane diffusivity using the converging wave method.

Keywords Anisotropy · Converging wave · Intermediate thickness · Thermal diffusivity

List of symbols

A Ratio of sample thickness to detection distance
I_0 Modified Bessel function of order zero
l Sample thickness

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N	Number of spatial dimensions
p	Dimensional parameter
Q	Heat source strength
r	Distance from center of the heat source
R	Radius of the heat source
t	Time
$t_{\max}$	Time at which thermal signal reaches maximum
T	Temperature

Greek symbols

α	Thermal diffusivity
α_r	Thermal diffusivity parallel to the surface
α_z	Thermal diffusivity perpendicular to the surface
ν	Coefficient of heat loss
$\tau_{\max}$	The minimum value of $t_{\max}$

1 Introduction

The converging thermal wave flash technique, originated by Cielo et al. [1], is used to measure the thermal diffusivity parallel to the surface of a sample. An instantaneous annular heat source is created on a flat surface using a laser pulse, which is passed through an axicon lens. The resulting temperature that rise as a function of time is recorded at the center of the annulus using a blackbody infrared (IR) detector (Fig. 1).

The transient thermal signal, $T(t)$, is described analytically in the study of Cielo et al. by

$$T(t) = \frac{Q}{(4\pi\alpha t)^p} \exp\left[-\frac{R^2}{4\alpha t}\right], \tag{1}$$

where Q is the strength of the source, α is the thermal diffusivity, and R is the radius of the annulus. The value of the parameter p in this equation is $N/2$, where N is the number of dimensions, and p is 3/2 if the sample is sufficiently thick to be described three-dimensionally, and is 1 if the sample is sufficiently thin so as to be described by a two-dimensional (2-D) approximation. This expression is derived by Carslaw and

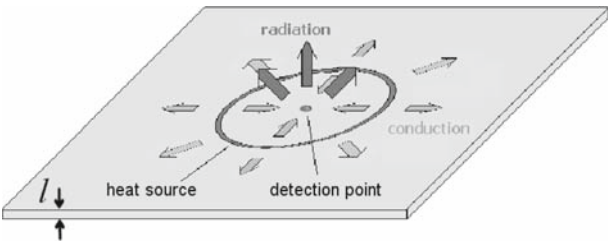

Fig. 1 An IR thermal transient is detected from the center of an instantaneous annular heat source

Jaeger [2]. By numerical curve-fitting Eq. 1 to the recorded thermal signal, α can be extracted. Alternatively, if the equation is differentiated, the time at which the transient reaches a maximum, $t_{\max}$, can be used to calculate α:

$$\alpha = \frac{R^2}{4pt_{\max}}. \tag{2}$$

An expanded form of Eq. 1 has been derived which takes account of the effect of heat losses and of non-centered signal detection [3]:

$$T(t) = \frac{Q}{(4\pi\alpha_r t)^p} \exp\left[-\frac{R^2 + r^2}{4\alpha_r t}\right] \exp\left[-\nu t\right] I_0\left(\frac{Rr}{2\alpha_r t}\right), \tag{3}$$

where α_r is the thermal diffusivity parallel to the surface, ν is the coefficient of heat loss, r is the off-center distance, and I_0 is the modified Bessel function of order zero.

The method has been used to measure the thermal diffusivity of thin sheets of copper, silver, aluminum, CVD diamond [4–6], nickel, and stainless steel [6]. Samples, which by their geometry fall between the two extremes of 'thick' and 'thin,' cannot be measured using Eqs. 1 or 3 unless an intermediate value of the dimensional parameter p can be assigned: $1 < p < 3/2$.

The range over which a sample can be described as thick or thin is given by the dimensionless parameter A,

$$A = \frac{l/\sqrt{\alpha_z}}{R/\sqrt{\alpha_r}}, \tag{4}$$

where l is the sample thickness (see Fig. 1). Equation 4 was derived by Enguehard et al. [7], who also derived analytically the limits defining thin and thick samples in terms of A. For thick samples, $A > 1.25$, for thin samples, $A < 0.3$, and for intermediate thickness samples, $0.3 < A < 1.25$.

In order to carry out a measurement on such an intermediate thickness sample, from an experimental viewpoint, it is possible to compensate for this by changing the radius of the annulus over a large range to make the sample 'thin' or 'thick' by comparison. However, this is not very convenient because if the annulus is too big, the signal at the center is greatly reduced, and if it is too small, errors due to the finite areas of heating and detection become significant. A better approach with intermediate thickness samples would be to assign a value of p in between its limits of 1 and 3/2, and using a version of Eq. 3 with this intermediate value of p, to find the thermal diffusivity by a curve-fitting algorithm.

An investigation of the intermediate thickness region was undertaken using finite-element (FE) modeling. Using the models of different thicknesses, the transition from thin to thick is evident in the change in the time at which the thermal signal reaches a maximum, $t_{\max}$, for models that otherwise use the same material properties and heat source size. A graph was made of the change of $t_{\max}$ with A, and a semi-analytical expression was fit to this. This can then be converted to a graph of the change of p with A, which is described by a similar semi-analytical expression. This allows an

intermediate value of p to be assigned, for any value of A, and measurements to be done on intermediate thickness samples.

In the case of anisotropic samples, using this semi-analytical equation, a method has been devised to obtain the ratio of anisotropy, α_r/α_z. If α_r is already known by the measurement of a thin or thick sample, then α_z can be obtained from the anisotropy ratio.

2 Theory

2.1 FE Model of the Thermal Signal

In order to ascertain how heat transfer varies in the case of thin and thick samples, FE models of an instantaneous ring-shaped heat source were generated on solids of different thicknesses.

In order to validate the model, parameters representing a laser beam heating a copper sample so that the heating surface has an annular shape of dimensions, 2.5 mm inner radius and 0.5 mm annular width, were used. The dimensions of the sample are ($15 \times 15 \times 0.1\,\mathrm{mm}^3$) (Fig. 2a), which classifies it as a thin sample and the coefficients representing the physical properties of copper (thermal conductivity of $401\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$, density of $8933\,\mathrm{kg \cdot m^{-3}}$, specific heat of $385\,\mathrm{J \cdot kg^{-1} \cdot K^{-1}}$) are used. The duration of the laser pulse (<10 ns) is infinitesimally small in comparison to the time-scale of the induced thermal transient (which reaches a maximum at about 20 ms), so it can be considered as an initial condition in the model.

The FE model was constructed using the software package ANSYS 5.6. The symmetry of the problem allows a 2-D analysis to be performed through a central section of the annulus (Fig. 2b). This section has a minimum depth of 0.1 mm and a width of 7.5 mm. This reduces the code run times considerably. The beam is absorbed within a depth of 100 nm (see very thin, top layer of elements in Fig. 2b) into the sample, and this region is known as the heated source. This is the depth to which the heat source has

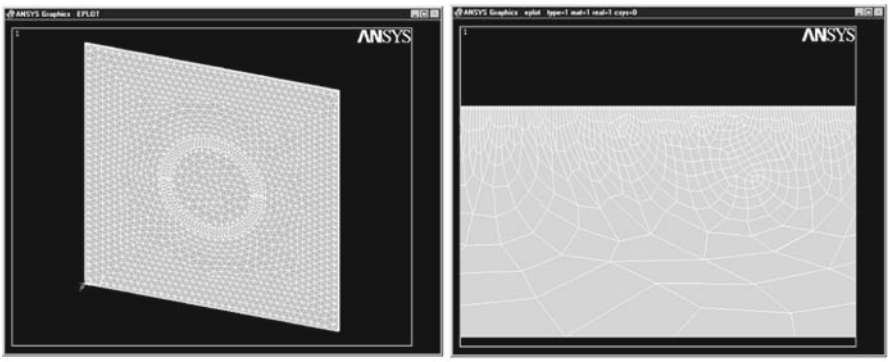

Fig. 2 Mesh used in ANSYS FE models of an annular heat source: (**a**) 3-D model of a thin sample and (**b**) 2-D model based on a cross section of the model in (**a**)

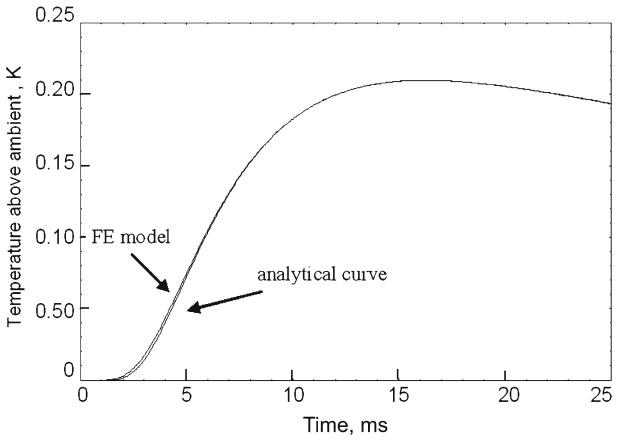

Fig. 3 Typical transient thermal signal generated by FE modeling overlaid with the analytical curve obtained from Eq. 2

spread by thermal conduction over the duration of the laser pulse. The laser radiation itself is fully absorbed by the solid within a few nanometers. This heated source is finely meshed. The remaining bulk area has increasing mesh coarseness with distance from the heated zone. A total of 1,787 elements are present in the mesh, and four-noded quadrilateral/triangular elements were used. The heat source was represented as an initial condition at the heating zone. Each node of the elements found in the heated zone was given an initial temperature of 45 K above ambient, which is the temperature rise of the volume of copper into which the heat has diffused over the period of the laser pulse. ANSYS settings were programmed to produce a transient model with a duration of 25 ms, in steps of 0.05 ms, which is a sufficient time for the thermal signal to reach a maximum and begin reducing again. The change of temperature with time at the node at the center point of the annulus was used to represent the thermal signal curve.

The thermal signal was seen to have excellent agreement with a curve generated from the analytical equation, without heat losses, provided by Cielo et al. [1] (Eq. 1, Fig. 3). The temperature at maximum time of the FE model curve was equal to 16.250 ms, compared to 16.256 ms, which is the time at maximum of the analytical curve calculated using Eq. 2, using a thermal diffusivity value of copper of $11.63 \times 10^{-5}\,\mathrm{m}^2 \cdot \mathrm{s}^{-1}$ [8] in each case.

2.2 Thin and Thick Samples

The high degree of correlation found between the model and the analytical expression motivated using it to examine the change of the parameter, p, with changing thickness. The model provided the flexibility of gradually increasing the depth of the sample until the thick sample regime was reached. The FE model accurately predicts the transient thermal signal for an annular heat source without the use of the dimensional approximations needed to extract the thermal diffusivity analytically or by curve-fitting (using Eqs. 1–3). The model can therefore be applied to any regime and not only to the 'thick'

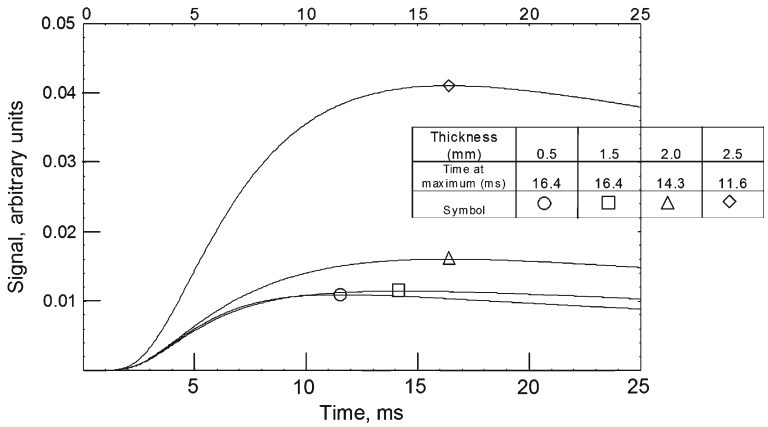

Fig. 4 Shift in the time at maximum (t_{max}) of the transient thermal signal as the thickness is varied for an isotropic sample

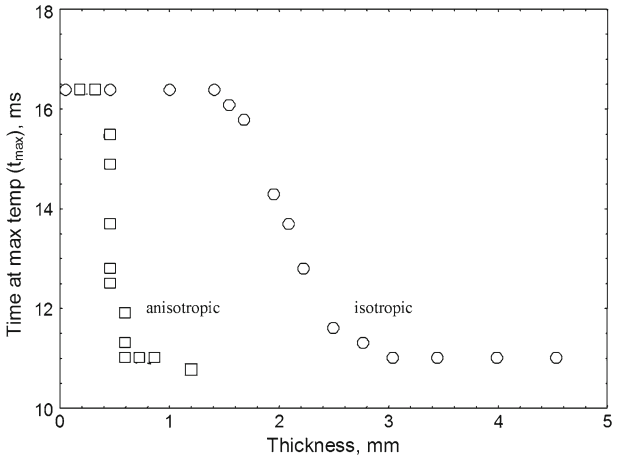

Fig. 5 Plot of t_{max} as the thickness is increased for isotropic (*circles*) and anisotropic samples (*squares*)

and 'thin' cases. Figure 4 shows the shift in the maximum of the thermal curve, t_{max}, with sample thickness.

The times at which the models of different thicknesses reach a maximum are graphed together in Fig. 5, for an isotropic material (circles), and for an anisotropic material (squares). It is clear how the maximum time varies between the two extremes of thin and thick samples. Below a certain thickness, about one-third the annular radius ($R = 2.75$ mm), heat conduction is approximately 2-D and all transients have the same peak time. Similarly, for thicknesses above about 1.25 times the annular radius, heat flow is fully 3-D and all transients have the same peak time, which is two-thirds that for the thin case.

In order to validate the method for an anisotropic material, a model was generated of a solid whose in-plane thermal conductivity was 20 times its perpendicular-to-plane

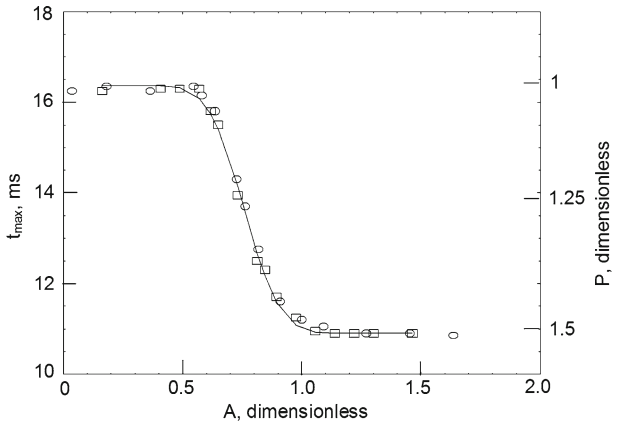

Fig. 6 Combined data sets of isotropic and anisotropic time maxima, in relation to the dimensionless parameter A, with a curve fit using the complementary error function. Isotropic results are represented by *circles*, and anisotropic results are by *squares*

thermal conductivity. The in-plane conductivity was set at the value of copper. This thermal anisotropy value is typical of layered materials such as graphite. A similar shift in the maximum time was observed as the sample was altered from being thermally thin to thermally thick, which is represented by squares in Fig. 5.

If the horizontal axis is converted to the dimensionless parameter A, using Eq. 4, then both sets of data points come together as shown in Fig. 6. From this graph, it can be seen that the intermediate thickness region can be described as being between the limits $0.3 < A < 1.25$ and that the change from thermally thin to thick is the same for isotropic and for anisotropic materials (and if the in-plane diffusivity is the same in each case, at the same value of $t_{\max}$).

A curve can be fit to the combined set of data points shown in Fig. 6, using the complementary error function. The curve has the general form,

$$t_{\max} = \tau_{\max} + \frac{\tau_{\max}}{4} \operatorname{erfc}\left[6A - 4.5\right], \tag{5}$$

where $\tau_{\max}$ is the minimum value of $t_{\max}$ on the curve. The specific curve shown in Fig. 6 has the formula,

$$t_{\max} = 10.9 + 2.73784 \operatorname{erfc}\left(6.01507A - 4.56634\right). \tag{6}$$

The curve fit is chosen solely to fit the shape of the data curve, rather than being based on any analytical model of the heat conduction in this case.

The vertical scale of the graph can be converted from $t_{\max}$ in ms, to p, which is a dimensionless unit, using a version of Eq. 2;

$$\frac{1}{p} = t_{\max} \frac{4\alpha_r}{R^2}, \tag{7}$$

where in this case, R, the radius of the annulus, is 2.75 mm, and α_r, the in-plane diffusivity, is $11.63 \times 10^{-5}\ \mathrm{m}^2 \cdot \mathrm{s}^{-1}$, in which case Eq. 6 becomes

$$\frac{1}{p} = 0.67223 + 0.16885\ \mathrm{erfc}(6.01507A - 4.56634). \tag{8}$$

When $t_{\max}$ is 10.9 ms, the sample is thick, and the value of $1/p$ should be exactly 2/3. Also, when the sample is thin, the value of p is exactly 1, and the value of $t_{\max}$ is 16.4 ms. Since the limits of the value of the $\mathrm{erfc}(x)$ are

$\mathrm{erfc}(x) = 0$ when x is large and positive ("thick sample") and
$\mathrm{erfc}(x) = 2$ when x is large and negative ("thin sample"),

then it is reasonable to simplify Eq. 8 to

$$\frac{1}{p} = \frac{2}{3} + \frac{1}{6}\mathrm{erfc}\,(6.01507A - 4.56634)\,, \tag{9}$$

so that for the thick and thin extremes,

$$\frac{1}{p} = \frac{2}{3} + \frac{1}{6} \times 0 = \frac{2}{3} \text{ and } \frac{1}{p} = \frac{2}{3} + \frac{1}{6} \times 2 = 1, \text{ respectively.}$$

If the argument of the complementary error function is simplified also, then the equation becomes

$$\frac{1}{p} = \frac{2}{3} + \frac{1}{6}\mathrm{erfc}\,(6A - 4.5)\,. \tag{10}$$

This is the general expression of the curve in Fig. 6, in terms of p. Using this expression, it is possible to calculate from the graph what the value of p should be, if the value of A is known, for any sample of intermediate thickness. Using this value of p, it is then possible to obtain the thermal diffusivity of such an intermediate thickness sample by inserting it in Eq. 3.

Also using this general expression, Eq. 10, it is possible to work backwards, using a set of time maxima from the measurements of varying thicknesses or annular radii, to get a value for A. Knowing A, a value for the ratio of anisotropy α_r/α_z can be obtained. To do this, a set of $t_{\max}$ data is recorded from the samples of varying thicknesses or measurements made with different radius annuli. Then a curve fit is made of the generalized expression, Eq. 5, to this set of data. The steps of the method are shown in Fig. 7.

3 Experimental

A typical experimental configuration is shown in Fig. 8. A short-pulse laser impinges on the sample to create an instantaneous heat source. The laser beam is first shaped into an annulus by an axicon lens. The thermal signal at the center of the annulus is transmitted to an IR detector using an optical fiber. The heat source is a Nd:YAG laser

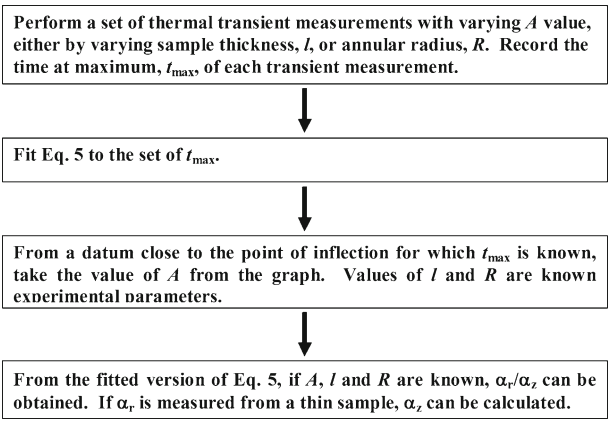

Fig. 7 Steps used to obtain the anisotropy of diffusivity

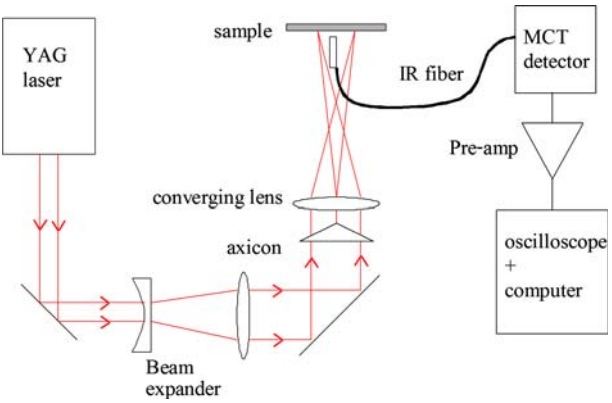

Fig. 8 Apparatus used to make converging thermal wave measurements

with a pulse length of 8 ns and a wavelength of 1064 nm. This pulse length ensures that the heat source is effectively instantaneous in comparison with the time scale of the transient produced, which typically reaches a maximum in 10 ms to 100 ms. The beam width is approximately 6 mm, and it is expanded by a factor of 2.5 through a Galilean beam expander. It's then converted to an annulus by a combination of an axicon (lens of conical section) and a converging lens. To access the front of the sample (the side that is illuminated by the laser), the optical fiber is made to cross the expanded laser beam, creating a small gap in the annulus, but this does not affect the recorded thermal signal.

The fiber is held at about 400 μm from the surface. The fiber is made from a polycrystalline silver halide material, which transmits in the wavelength range of 4 μm to 18 μm. It's directly pig-tailed to a 1 mm^2 sensitive element in a HgCdTe detector, and the fiber core diameter is 900 μm. The signal is amplified in a pre-amp and then passes to an oscilloscope, where it is digitized. To improve the signal-to-noise ratio, several hundred measurements are taken in quick succession and these are averaged

on the oscilloscope, before being imported onto a pc spreadsheet. The firing of the laser and the collection and averaging of the signal by the oscilloscope are controlled by a computer using Labview software.

4 Results

4.1 Isotropic Samples of Intermediate Thickness

In order to test the correspondence of the complementary error function curve, Eq. 5, to a set of t_{max} values for an isotropic material, measurements were made on copper samples of thicknesses of 0.15 mm, 1 mm, 2 mm, 3 mm, and 5 mm, with an annulus of radius of 4.2 mm. The t_{max} value of each recorded transient was obtained by fitting a third-order polynomial curve to the data with Mathematica software. A typical set of data with a polynomial curve fit is shown in Fig. 9. The maximum point of the curve is then selected as t_{max}.

A graph was made of all t_{max} values as a function of A, and using a nonlinear curve fitting routine in Mathematica, the generalized Eq. 5 was fit to the set of t_{max} values, producing the specific curve in the case of copper shown in Fig. 10, with the formula,

$$t_{max} = 18.4363 + 4.60907\text{erfc}\,[6A - 4.5], \tag{11}$$

where the value of τ_{max} is 18.4363 ms. This value is greater than obtained in the FE simulations (Fig. 6, Eq.6) due to the greater annular radius used.

There is good correlation between the curve and the data points, which confirms the form of the expression generated from the FE modeling results.

By changing the vertical scale of the graph shown in Fig. 10, so that the maximum of the curve corresponds to $p = 1$ and the minimum of the curve corresponds to $p = 3/2$, a value of p can be assigned to samples which lie between the thin and thick extremes. This value of p can be inserted in Eq. 3, and using nonlinear curve-fitting

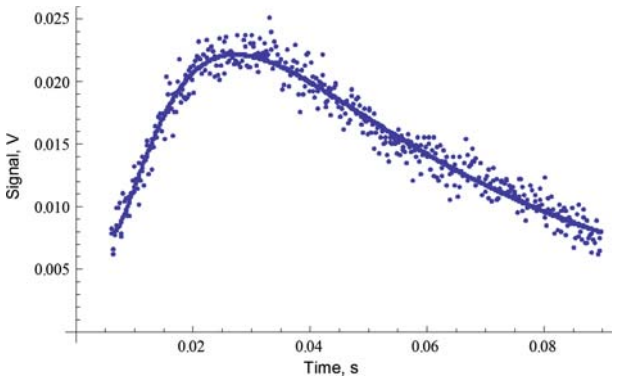

Fig. 9 Recorded transient and polynomial curve fit to find t_{max} for Cu sample of 2 mm thickness

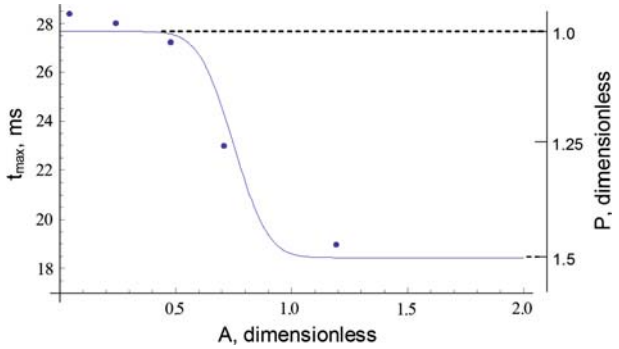

Fig. 10 Curve fit of Eq. 5 to the set of $t_{\max}$ values for copper

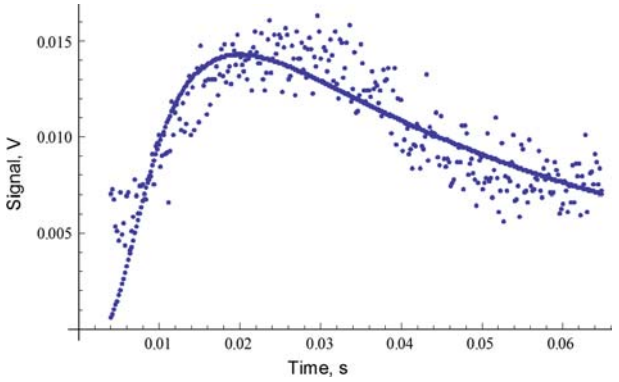

Fig. 11 Curve fit used to obtain the thermal diffusivity of an intermediate thickness sample of copper with $p = 1$

algorithms described elsewhere [3], a thermal-diffusivity value can be extracted for that sample.

For example, for the sample of 3 mm thickness, $A = 0.714$ and from Fig. 10, p has the value of 1.2. Inserting this in Eq. 3,

$$T(t) = \frac{Q}{(4\pi\alpha_r t)^{1.2}} \exp\left[-\frac{R^2 + r^2}{4\alpha_r t}\right] \exp\left[-\nu t\right] I_0\left(\frac{Rr}{2\alpha_r t}\right), \tag{12}$$

and numerically fitting the curve to the measured signal, the graph shown in Fig. 11 is obtained, resulting in the value for the thermal diffusivity of $(11.7 \pm 2.8) \times 10^{-5}\ \mathrm{m}^2 \cdot \mathrm{s}^{-1}$, which is close to the accepted value: $11.63 \times 10^{-5}\mathrm{m}^2 \cdot \mathrm{s}^{-1}$ [8].

Similarly, a measurement was performed on a sample of iron of 3 mm thickness. Using the value of $p = 1.2$ inserted in the curve-fitting Eq. 3, a value for the thermal diffusivity of $2.1 \times 10^{-5}\ \mathrm{m}^2 \cdot \mathrm{s}^{-1}$ was obtained, which is close to the accepted value of $2.27 \times 10^{-5}\ \mathrm{m}^2 \cdot \mathrm{s}^{-1}$ [8].

Table 1 Thermal diffusivity of anisotropic graphite sample, measured perpendicular (α_z) and parallel (α_r) to the surface

	Thickness (μm)	Thermal diffusivity	
		Direction	(10^{-5} m$^2 \cdot$ s^{-1})
Conventional flash method	844	α_z	0.69
Converging wave, thin sample	255	α_r	20.9
Converging wave, thick sample	844	α_r	20.1

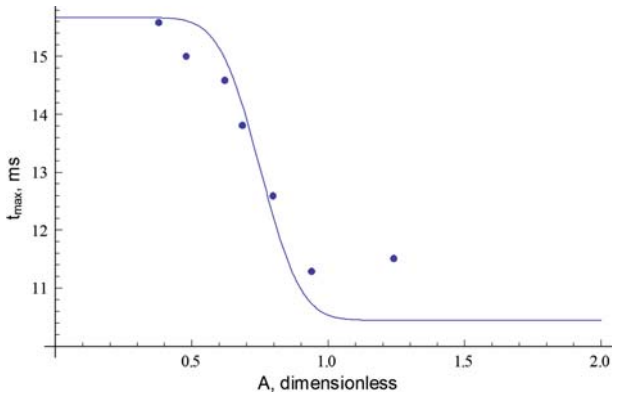

Fig. 12 Curve fit of Eq. 5 to the set of $t_{\max}$ values for graphite

4.2 Anisotropic Sample

A graphite sample was used to test the agreement of the complementary error function curve, Eq. 5, with a set of $t_{\max}$ values, for an anisotropic material. The thermal diffusivity of this material was first measured perpendicular to the plane (α_z), using the conventional laser flash method [9] and parallel to the plane (α_r), in thin and thick extremes (at thicknesses of 255 μm and 844 μm), using the converging wave method (Table 1).

Samples were machined to several thicknesses between 844 μm and 255 μm, and converging wave measurements were made on them, and the value of $t_{\max}$ was obtained for each transient. To make a curve fit with the erfc expression, Eq. 5, it was necessary to first convert the thicknesses l to values of A, using Eq. 4. For this, an estimated value of $\sqrt{\alpha_r}/R\sqrt{\alpha_z}$ was used, based on the thermal-diffusivity values in Table 1. The erfc curve subsequently obtained as a fit to the set of $t_{\max}$ values is shown in Fig. 12. The conversion of variables is shown in Table 2.

There is good agreement between the set of $t_{\max}$ values for graphite and the curve, which has the form,

$$t_{\max} = 10.45 + 2.61\mathrm{erfc}\,[6A - 4.5]\,, \tag{13}$$

Table 2 Values of A and t_{max} used for curve fit of anisotropic graphite

Thickness (l) (μm)	$\left(\sqrt{\alpha_r}/R\sqrt{\alpha_z}\right) = 1470\,\mathrm{m}^{-1}$	
	$A = l \times \left(\sqrt{\alpha_r}/R\sqrt{\alpha_z}\right)$	t_{max} (ms)
255	0.375	15.6
325	0.478	15.0
423	0.622	14.6
465	0.684	13.8
542	0.797	12.6
638	0.938	11.3
844	1.241	11.5

where $t_{max} = 10.45$ ms. This confirms that the general expression, Eq. 5, is valid for anisotropic samples.

In this study, the curve fit of Eq. 5 to the set of t_{max} values for graphite was not used directly to calculate the thermal anisotropy ratio α_r/α_z. This requires weighted curve-fitting analysis software, which can perform a curve fit when the horizontal axis (value A) is not fixed, and this was not available to the authors at the time of the investigation. However, the relationship between the set of time maxima, t_{max} values, and the thermal thickness to heat source radius, A, can be seen in Fig. 12, and this may be used to calculate the thermal anisotropy, α_r/α_z, using the appropriate numerical curve-fitting software.

5 Conclusions

An expression has been derived, based on the results of FE numerical simulations, to describe the change of the transient thermal signal which is produced in the converging wave method for measuring the thermal diffusivity, as the sample thickness is changed from thermally thin to thick. This expression has been verified by fitting it to experimental results obtained from isotropic samples of copper and iron, and anisotropic graphite. The expression can be used to assign an intermediate dimensional parameter, p, to a sample which falls between the extremes of thin, or 2-D heat flow, and thick, or 3-D heat flow and using this value, the thermal diffusivity of such a sample can be measured by the converging wave method. This extends the range of the method considerably and avoids the need to machine the samples to required thicknesses or to change the radius of the annular heat source to carry out measurements in the intermediate thickness range. The expression can also be applied to anisotropic materials, and a method is described to extract the ratio of the in-plane to perpendicular-to-plane thermal diffusivity.

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