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Development of an algorithm to extract thermal diffusivity for the radial converging wave technique

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Abstract

The usual equation for the converging wave method [P. Cielo, L.A. Utracki, M. Lamontagne, Thermal diffusivity measurements by the converging thermal-wave technique, Canad. J. Phys. 64 (1986) 1172–1177] for thermal diffusivity measurements assumes idealised conditions that are difficult to achieve in a real experimental situation and this hinders the extraction of diffusivity values. A model for thermal transport is described here that takes into account errors due to heat losses and is relatively insensitive to detection position inaccuracy. A simple polynomial equation is derived from the model and it is used to generate initial guesses for the Levenberg–Marquardt algorithm, which uses these initial guesses to avoid a local minimum problem and ultimately produces a value for radial thermal diffusivity.

Keywords: Thermal diffusivity; Flash method; Converging wave; Heat loss corrections; Curve-fitting

1. Introduction

The converging wave technique developed by Cielo et al. [1] is a development of the Parker method [2–4], which measures the radial thermal diffusivity of solids. The flash method of Parker is one of the most reliable methods of measuring thermal diffusivity. However, it is limited to measuring diffusivity in the direction perpendicular to the plane, of relatively thin, planar samples. In the Cielo method, a planar sample is subjected to a very short pulse of annular shaped radiant energy from a laser and the resulting temperature rise at the centre is determined using an IR detector. By recording the time resolved temperature, thermal diffusivity values can be derived. The geometry allows diffusivity to be measured parallel to the plane, for samples of potentially any thickness.

The use of the converging thermal wave analysis to determine radial thermal diffusivity has been described by current literature [1,5–7]. However it can only be applied under adiabatic and ideal experimental conditions that are difficult to fulfil in practice. Two of the fundamental assumptions are centre-point detection and that there is no heat loss by radiation and convection. In a similar study the sample was placed in a vacuum in order to eliminate convection loss [8]. In reality there may well be an error in the centring of the system. In addition this study shows that heat loss mechanisms,

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Nomenclature

c	specific heat
F	radiating surface of body
H	coefficient of convective surface heat transfer
I_0	Modified Bessel function of order zero
r	off centre distance
r'	radius of the annulus
t	time
T	temperature
T_0	ambient temperature

Greek symbols

α	thermal diffusivity
ε	emissivity
ν	heat loss term
ρ	density
σ	Stefan–Boltzmann constant
$Q\rho c$	energy absorbed by the sample

particularly radiation, must be taken into account if a good match between theory and experiment is to be made.

Data reduction methods play a significant role in determination of the thermal diffusivity. In the original paper published by Cielo, based on the converging wave technique, only the time at maximum temperature or the time at half the maximum temperature is taken from the measured temperature versus time curve and used to calculate the thermal diffusivity. In a paper written by Kim et al. [5] the times at which the curve reaches a certain equal temperature, in the ascending and descending temperature evolution curve, t_1 and t_2, say, were used to determine the thermal diffusivity. Using the Kim method it was necessary to know the material being measured in advance because the amplitude at which t_1 and t_2 are taken is subject to the type of material being measured. Kim's method was a good test to see if the experimental data for a known sample produced results that were within an acceptable range of quoted values in literature, but for unknown samples the choice of amplitude may be difficult to ascertain. In addition both methods do not use the whole dataset of experimental results particularly where departures from the idealised conditions become apparent. The progress in computers over the last number of years has ensured that curve-fitting algorithms can use all data points simultaneously to extract the required parameters. This ensures that non-linear curve fitting routines can be used on converging wave data on realistic timescales. In general it has been written, "*The theoretical and numerical analysis required for obtaining thermal diffusivity are one of the challenges of the photothermal radiometry technique for arbitrary specimen shapes and heating beam geometries*" [9].

This paper develops the mathematical formulism that is necessary for describing real data in the converging wave method by taking account of heat loss and errors in centring. This is then used in a curve-fitting method on the experimental data and the results found for this method correlate very well with values found in the literature for various materials (see Table 1). The model equation used in the curve-fitting process has been developed so it can account for non-adiabatic conditions and it can also account for centring limitations based on the detectors used in the experiment. The mathematical model still assumes that an instantaneous heat pulse is absorbed in a very thin layer of the sample and the laser beam impinging on the sample has a uniform annular shape. These approximations are shown to be valid in our measurements.

To guarantee good measurements it is very important to accurately measure the annular radius. This is achieved using a software technique whereby a CCD camera captures an arc of the annulus. To find the median ring of the annulus, image-processing algorithms are applied. The data provided from these algorithms provide the coordinates for the arc. This data is then implemented into a multiple linear regression algorithm to ultimately determine the centre point and the mean radius of the annulus.

Table 1
Results on three well-characterised materials, compared with reference values and the conventional laser flash technique

Material	Thermal diffusivity $\times 10^{-5}\,m^2\,s^{-1}$				
	Reference—CRC [18]	Measured radial	Agreement radial and ref (%)	Measured laser flash	Agreement radial and flash (%)
Copper	11.625	11.71	1	11.60	1
Aluminium	9.538	9.49	1	8.93	6
Zinc	4.187	4.07	3	4.38	8

The same samples were measured using a version of Parker's flash method described elsewhere [10–12], and the results got by this method also compare well with those obtained using the radial method and curve-fitting procedure [Table 1].

2. Mathematical theory

The ideal model [1] is based on the behaviour of a homogeneous, thermally insulated, infinite slab with an instantaneous heat source uniformly applied over the front surface of the sample. A typical experimental configuration is shown in Fig. 1 where a short pulse laser (typically < 10 ns) impinges on the sample. The beam is shaped into an annulus and an infrared detector measures the thermal signature on the back surface of the sample at the centre. The temperature rise found at the back surface of a sample is less that 10 K. This ensures that the infrared detector taking measurements has an error, of less than 0.5% when extracting thermal diffusivity for the laser flash method. This is mainly due to a small non-linearity given by Planck's law [13,14].

The partial differential equation that is used to describe the converging wave technique mathematically is developed from the equation for an instantaneous point source of heat, that is, a finite quantity of heat liberated at a given point (x', y', z').

Therefore the PDE for the conduction of heat from a point source to a location (x, y, z) can be written as [15]:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = \frac{1}{\alpha}\frac{\partial T}{\partial t} \quad (1)$$

and is satisfied by,

$$T = \frac{Q}{(4\pi\alpha t)^{3/2}} \exp\left[-\frac{(x-x')^2 + (y-y')^2 + (z-z')^2}{4\alpha t}\right] \quad (2)$$

The heat liberated is $Q\rho c$.

Samples on which measurements were performed were metal foils, which were sufficiently thin to approximate to a two dimensional case [16]. For a two-dimensional sample, in the xy-plane, the following holds:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} = \frac{1}{\alpha}\frac{\partial T}{\partial t} \quad (3)$$

and the solution which describes the temperature at a point (x, y) due to a point source at (x', y') is

$$T = \frac{Q}{(4\pi\alpha t)} \exp\left[-\frac{(x-x')^2 + (y-y')^2}{4\alpha t}\right] \quad (4)$$

2.1. Derivation of the heat loss term

The heat loss term is a very significant factor in terms of non-ideal departures from the ideal equation. A mathematical derivation of heat losses in the experiment is based on a combination of convective and radiative heat losses (Fig. 2), which are described by Newton's law of cooling and the Stefan–Boltzmann expression respectively. Addition of the heat loss term in Eq. (1) is achieved by examining the Stefan–Boltzmann expression and Newton's law of cooling as a linear component to the P.D.E equation.

The Stefan–Boltzmann expression can be simplified since the temperature rises only a few degrees above ambient at the centre of the annulus, i.e. $T - T_0$ is small.

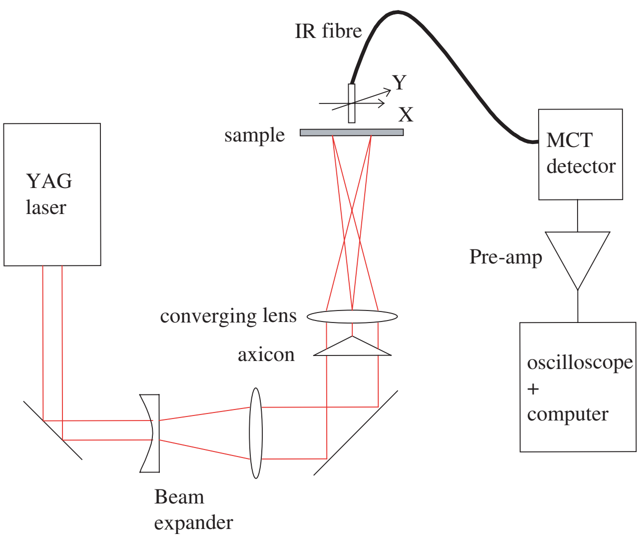

Fig. 1. Experimental configuration for the radial thermal diffusivity set-up.

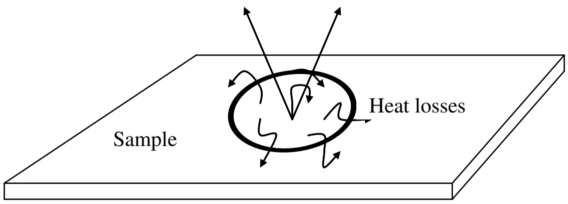

Fig. 2. Shows the heat losses from the surface of the sample.

This physical phenomenon mathematically reduces the Stefan–Boltzmann expression into a linear component by using the Taylor expansion and simplifying. Eq. (5) shows the simplification of the expression.

$$\text{Radiative heat loss} = \sigma\varepsilon F(T^4 - T_0^4) \approx 4\sigma\varepsilon F T_0^3 (T - T_0) \quad (5)$$

Newton's law of cooling is used to describe heat loss due to forced convection as

$$\text{Convective heat loss} = H(T - T_0) \quad (6)$$

Since Newton's law of cooling has a linear component, both heat loss terms can be combined as in Eq. (7). Total losses are

$$4\sigma\varepsilon F T_0^3 (T - T_0) + H(T - T_0) = v(T - T_0) \quad \text{where } v = H + 4\sigma\varepsilon F T_0^3 \quad (7)$$

The heat loss term can be added to the PDE equation (1) and is written as:

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = \frac{1}{\alpha}\left(\frac{\partial T}{\partial t} + v(T - T_0)\right) \quad (8)$$

It can be shown [17] that Eq. (9) is a solution to Eq. (8).

$$T(t) = \frac{Q}{(4\pi\alpha t)^{3/2}} \exp\left[-\frac{x^2 + y^2 + z^2}{4\alpha t}\right] \exp[-vt] + T_0 \quad (9)$$

Since the actual temperature is not measured, and only the change in the blackbody signal due to the heat source is recorded, ambient temperature, T_0, can be given the value zero.

2.2. Combining the mathematical expression for heat losses and off-centre detection

The solution for the point source is a key feature for the ultimate mathematical formulism required for the converging wave technique, which can be developed from this. From this solution of the point source the geometry for the annular laser heat source can be generated. The mathematical geometry for the ring source is found by first converting the Cartesian coordinates to polar for the point source. This can be described mathematically by the cosine rule (Eq. (10), see Fig. 3). An off

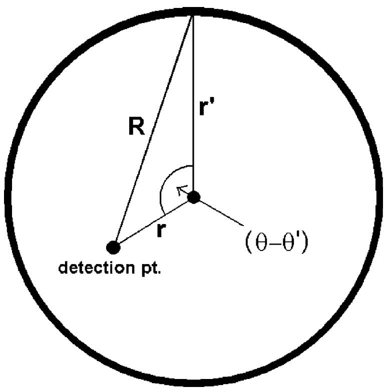

Fig. 3. Describes the geometry of the off-centre component on the ring.

centre component, r, is then included in the polar coordinate description.

$$R^2 = (x - x')^2 + (y - y')^2 = r^2 + r'^2 - 2rr'\cos(\theta - \theta') \quad (10)$$

To create an instantaneous ring heat source, point sources of strength $Qr'\mathrm{d}\theta'$ are distributed around the circle $r = r'$. The temperature at time t at the point whose polar coordinates are (r, θ) is

$$\frac{Qr'}{(4\pi\alpha t)^{3/2}} \int_0^{2\pi} \left\{\exp\left[-\frac{r^2 + z^2 + r'^2 - 2rr'\cos(\theta - \theta')}{4\alpha t}\right] \times \exp[-vt]\right\} \mathrm{d}\theta' = \frac{Q'}{(4\pi\alpha t)^{3/2}} \exp\left[-\frac{r^2 + r'^2 + z^2}{4\alpha t}\right] \exp[-vt] I_0\left(\frac{rr'}{2\alpha t}\right) \quad (11)$$

where $Q' = 2\pi r'Q$, and the total quantity of heat absorbed by the sample is $Q'\rho c$. I_0 is the modified Bessel function of order zero. Eq. (11) describes the case for a continuous ring source with heat losses and there is also an extra parameter r included which describes the off-centre detection. This equation can be simplified if detection is perfectly centred i.e. if the off centre distance was equal to zero and this approximation has been made previously [1,5–7]. But in reality, some off-centre detection is difficult to avoid.

For a thin sample, heat-flow can be assumed to be overwhelmingly two-dimensional and can be described by the mathematical extreme of heat flow in the xy-plane. The expression then becomes

$$T = \frac{Q'}{(4\pi\alpha t)} \exp\left[-\frac{r^2 + r'^2}{4\alpha t}\right] \exp[-vt] I_0\left(\frac{rr'}{2\alpha t}\right) \quad (12)$$

Other geometrical sources of error are the finite detection area and the finite thickness of the heat source.

The expression as derived describes the temperature at a single point near the centre of a ring of infinitesimally small point sources. Previously it has been assumed that if the detection area and source thickness are less than about one tenth of the source radius, they have a negligible effect on the thermal transient [7]. Finite element models were generated with non-zero source and detection areas and found to agree with this.

Eq. (12) can be simplified using the coefficients A, B and C:

$$A = \frac{Q'}{4\pi\alpha}, \quad B = \frac{r^2 + r'^2}{4\alpha}, \quad C = \frac{rr'}{2\alpha}$$

and it is rewritten as:

$$T(t) = \frac{A}{t}\exp\left[-\frac{B}{t}\right]\exp[-vt]I_0\left(\frac{C}{t}\right) \tag{13}$$

In summary, Eq. (13) is used to account for heat losses in the experiment as well as taking off-centre signal due to misalignment into the analysis. This mathematical treatment is necessary since experimental deviations from the simple mathematical formulism are satisfied by these two physical properties being included in the mathematical analysis.

3. Data reduction algorithms

3.1. Curve-fitting procedure

In order to extract values of thermal diffusivity a curve-fitting algorithm must be used in conjunction with Eq. (13). In this study the Levenberg–Marquardt algorithm is used. However care must be taken to avoid trapping the algorithm in local minima. Careful choice of starting values are necessary, as the parameter estimates used by the Levenberg–Marquardt algorithm may represent a *local minimum* in the sum of squared residuals $\sum_i e_i^2$.

Initial guesses for the parameters can be found by a polynomial fit on the experimental data and this has been found to be a reliable starting point for the full curve-fitting algorithm. In order to fulfil this criterion equation (13) needs to be simplified into a product of exponentials and this is achieved by approximating the Bessel factor to an exponential term. The power series for the exponential and Bessel terms are very similar over a range of values for the arguments of both terms:

$$I_0\left[0, \frac{1}{t}\right] = \sum\left[\frac{t^{-2n}}{(2^{2n})(n!)^2}\right] \tag{14}$$

$$\exp\left[\frac{1}{t^2}\right] = \sum\left[\frac{t^{-2n}}{(n!)}\right] \tag{15}$$

and an approximate substitution can be made:

$$I_0\left(\frac{C}{t}\right) \approx \exp\left(\frac{D}{t^2}\right) \tag{16}$$

where $D = \frac{C^2}{4}$.

The exponential approximation for the Bessel function as described in Eq. (16) is satisfied for a range of C values. This range of values was found by fitting the Bessel and exponential terms respectively, until they no longer agreed with the condition described in Eq. (16). The range of values for which the parameters C and D satisfy the condition in Eq. (16) are $0.0001 < C < 0.1$ and $2.5 \times 10^{-9} < D < 2.5 \times 10^{-3}$. These values found for the parameters C and D over which the approximation is valid, correspond to thermal diffusivity for nearly all known solid materials and for a transient time recorded up to 1 s. For example the off centre detection distance is usually 0.25 mm and the radius of the annulus is given as 3.3 mm. Thus the substitution for the Bessel function holds for a range of thermal diffusivities from $4 \times 10^{-6}\,\mathrm{m^2\,s^{-1}}$ to $4 \times 10^{-3}\,\mathrm{m^2\,s^{-1}}$.

Using the exponential substitution for the Bessel function equation (13) becomes.

$$T(t) \approx \frac{A}{t}\exp\left[-\frac{B}{t}\right]\exp[-vt]\exp\left[\frac{D}{t^2}\right] \tag{17}$$

The natural logarithm is then applied along Eq. (17) and this has the effect of simplifying the equation to a polynomial form.

$$\log_e tT(t) \approx \log_e(A) - \frac{B}{t} - vt + \frac{D}{t^2} \tag{18}$$

The polynomial-fitting procedure is then used to fit this equation to the data to generate initial guesses for the four parameters for the Levenberg–Marquardt algorithm. The parameters can be interpreted physically as:

A	Amplitude of signal
B	Term for finding diffusivity
D	Off-centre component
v	Heat loss parameter

The extracted parameters from the polynomial fit constitute initial guesses for the non-linear fit. These initial guesses are then used in the Levenberg–Marquardt algorithm. This algorithm revises the initial guesses using the sum of squared residuals. The final outcome of the algorithm provides the user with the four fitted parameters. The most important parameter is the B parameter, which provides the user with the thermal diffusivity.

3.2. Determination of the radius of the annulus

In order to determine the radius of the annulus, the active element of a CCD camera was substituted for

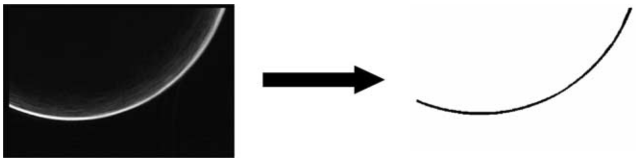

Fig. 4. The mean path of the beam profile is shown at the right.

the sample and the laser attenuated. Since the annulus was larger than the CCD element, only part of the annulus was captured. An alternative would have been to use image reduction optics but this could have introduced errors if the demagnification was not known accurately.

Fig. 4 shows a typical captured arc. The (x_i, y_i) coordinates for the arc are extracted using image-processing algorithms. These image-processing algorithms are based on digitising the bitmap image so that the coordinate profile for the annular beam can be stored. The coordinates for the beam are then analysed using a multiple linear regression.

The multiple linear algorithm is designed to extract the radius and centre point of the circle from the annular arc. The implementation of the algorithm requires that the equation of a circle be parameterised and the centre point of the circle be defined as (x_0, y_0). The parameterisation for the equation of a circle leads to the following substitutions. Defining

$$z = x^2 + y^2, \quad b = -2x_0, \quad c = -2y_0, \quad a = x_0^2 + y_0^2 - r^2$$

the circle becomes

$$z + bx + cy + a = 0 \tag{19}$$

Multiple linear regression is used to fit a circle centre (x_0, y_0) and radius r to the observed annular arc. Thus we wish to minimise

$$\Pi = \sum_{i=1}^{n} [z_i + a + bx_i + cy_i]^2 \tag{20}$$

Note that a, b, and c are unknown constants while all x_i, y_i are given from the measured arc. To obtain the least squares fit the function Π is differentiated for a, b, and c and their expressions are set equal to zero, so as to extract the minimum value. Finally the unknown coefficients a, b and c can then be obtained by solving the linear equations and a value for the radius and centre point (x_0, y_0) can be obtained.

4. Results and discussion

In order to demonstrate the improvement in fitting experimental data to the theoretical expressions a typical time resolved measurement for copper is shown in Fig. 5 together with a fitted curve of the form of Eq. (4), that is without taking account of heat losses and non-centred detection.

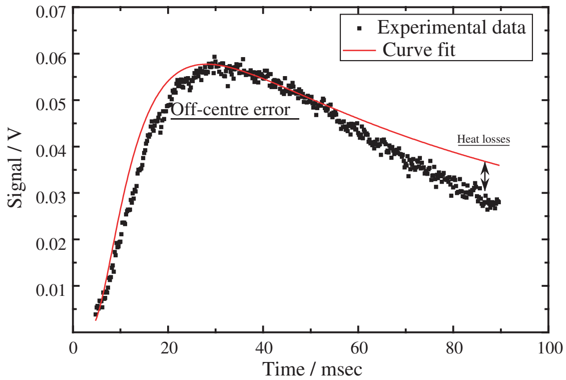

Fig. 5. Curve-fit using the simple, ideal expression.

Several features can be identified with the additional terms used in this study. Firstly the peak of the thermal transient is shifted due to off-centre detection. Essentially the ideal equation assumes that the components of the thermal wave generated by the annular heating arrive at the detection point simultaneously. This, in practical terms is difficult to guarantee. The second feature is the premature drop in temperature at the detection area due to the heat loss mechanisms described earlier, this also has the effect of shifting the maximum peak position of the thermal transient. Fig. 5 demonstrates that measurements using the ideal theory, that rely on determination of the time to reach the peak temperature or the time lapse between predetermined temperature values could introduce errors if these phenomena are not accounted for experimentally.

Fig. 6 shows the same experimental data with a curve fitted using the algorithm containing the correction terms in Eq. (13). The value for thermal diffusivity extracted from the fit is in good agreement with quoted values. In this case no subjective decision is used to determine which data points are used in the calculation and the complete data set is used.

When re-examining Eq. (13) it should be pointed out that the parameter B has a radius-squared term, that is the radius found for the annular beam needs to be highly accurate since the Levenberg–Marquardt fitting routine provides an empirical value for this parameter. The soft-

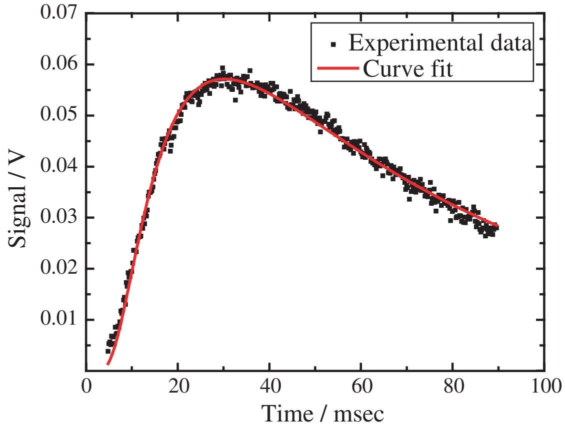

Fig. 6. Curve-fit using the expression developed to include effects of heat losses and non-centring.

ware approach described above shows the steps required for evaluating the radius and centre point of an annular arc. It also provides pixel accuracy and is more reliable than measuring the radius by hand. This method for measuring the radius can be applied when it is difficult or time consuming to form a perfect annular profile on a CCD camera. If a full annular beam is captured by the camera a direct measurement of the annulus can be made without the need of the circle fitting algorithms.

Table 1 shows diffusivity values obtained from three different metal foils, compared with values taken from literature [18] and compared with values obtained when the same samples were measured using a different technique, the conventional laser flash method. There is good agreement between all the values.

5. Conclusions

This paper describes the mathematics and algorithms required to account for non-ideal experimental conditions in the converging wave method of thermal diffusivity measurement. These investigations showed that the non-centred detection and the heat losses which occur naturally in the experiment were the two main causes of deviation from the ideal curve. Thus having understood the nature of the non-ideal conditions occurring in the experiment, a mathematical development of the ideal equation was used to account for these physical conditions.

An expression was used to account for the off-centre components, using a Bessel function solution. Comparison with experimental data showed that this could account for an apparent shift in the peak of the thermal transient resulting in a potential error in thermal diffusivity measurement.

Heat loss (by radiation and convection) was also shown to be a potential source of error in experimental measurements. It was shown that a Stefan–Boltzmann expression for radiation and a Newton's law of cooling term could be added to the ideal equation to describe these loss mechanisms.

The final derived equation was compared to experimental data using curve-fitting procedures. The main curve-fitting algorithm used was the Levenberg–Marquardt. Good initial guesses are required for optimal usage of the Levenberg–Marquardt algorithm. It was found that this is necessary since if the initial guesses provided are poor they can lead to a local minimum. The equation was arranged in polynomial form and a linear fit provided the initial guesses for input into the Levenberg–Marquardt algorithm. With this approach accurate values of thermal diffusivity were derived for various materials without using any subjective selection of the data set.

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