International Journal of Thermophysics, Vol. 11, No. 5, 1990

On the Analysis of the Thermal Diffusivity Measurement Method with Modulated Heat Input

R. De Coninck[1]

Received September 25, 1989

The present paper proposes a simplified way to analyze thermal diffusivity experiments in which the phase shift is measured between the modulations of the temperatures on either face of a disk-shaped sample. The direct application of complex numbers mathematics avoids the use of the cumbersome formulae which hitherto have hampered a wider confirmation of the method and which restricted the range of the phase lag to an angle of 180°. The algorithm exposed makes it more practical to refine the analysis, which may lead to a higher accuracy and a wider use of the method. The origins of some possible errors in the calculated results are briefly reviewed.

KEY WORDS: Complex numbers analysis; modulated heat input; phase shift measurement; thermal conductivity; thermal diffusivity.

1. INTRODUCTION

In 1960–1961 Cowan [1, 2] proposed a modification of the well-known Angström method for measuring thermal diffusivity at high temperatures on disk-shaped samples, the temperature modulation phase lag method. The development of this method and its associated mathematics marked a breakthrough in the field of thermal diffusivity-conductivity measurement techniques. Today it still is an efficient and up-to-date method next to the well-known pulse or flash method.

Cowan demonstrated the feasibility to calculate, from a phase lag measurement only, the thermal diffusivity and conductivity of a disk-shaped sample. One face of a thin disk of the material to be measured (see Fig. 1) is brought to a given equilibrium temperature by an adjustable

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[1] Materials Development Department, SCK/CEN, B-2400 Mol, Belgium.

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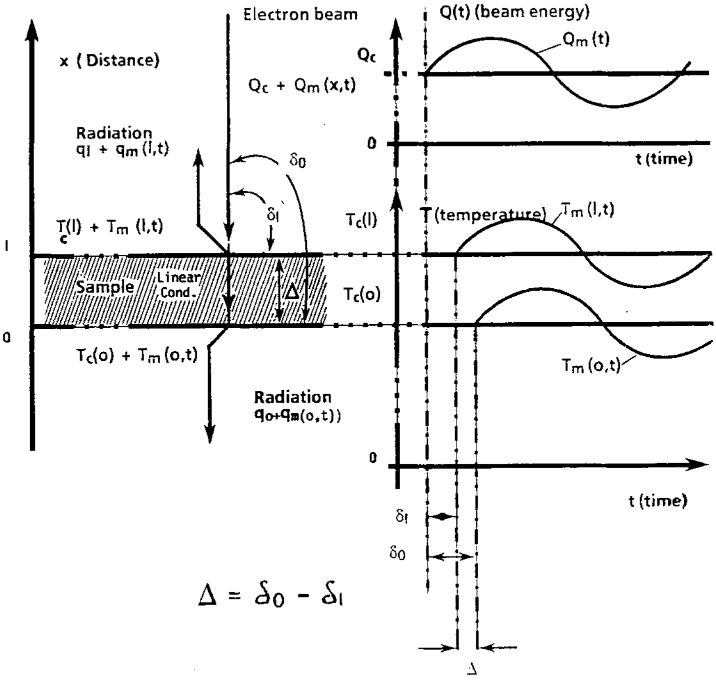

Fig. 1. Schematic experimental sample arrangement.

energy influx (e.g., thermal radiation, laser irradiation, electron beam bombardment, or the like) and this energy is sinusoidally modulated in amplitude.

Three phase shifts are to be distinguished: the shift δ_l between the heating beam and the temperature modulation of the heated surface, the shift δ_0 between the heating beam and the temperature of the nonheated surface, and the shift Δ over the sample. They are connected to each other by the relation[2]

$$\Delta = \delta_0 - \delta_l \tag{1}$$

Cowan showed that the measurement of Δ was to be preferred over δ_l or δ_0, particularly since delicate interfering heat loss parameters have minimum influence in that case. Also, the measurement of Δ can generally be carried out more easily and more accurately. Subsequent literature is in agreement with the statements of Cowan; consult, e.g., an exhaustive survey concerned with thermal modulation diffusivity methods [3].

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[2] For a complete definition of symbols refer to the Nomenclature.

Nevertheless, Cowan did not derive an explicit expression for Δ but only for δ_l and δ_0 separately, and these expressions are rather cumbersome. Credit should be given to Wheeler [4] for his progressive approach of the problem and his introduction of a practical iterative calculation method.

Only few authors have dealt with further developments of Cowan's original work. Cerceo and Childers [5] derived an equation which directly gives Δ but their analysis is valid for a special simplified condition only. Penninckx [6] presented two (in fact equivalent) direct and relatively simple equations for Δ which are consistent with Cowan's individual expressions for δ_l and δ_0.

In the recent years, a number of alternative possibilities were explored to determine thermal diffusivity, next to the "classical" flash and modulation methods. They have arisen from, among others, optoacoustic, photothermal, photodeflection, or thermoelastic disciplines. This certainly opened interesting horizons, especially since these methods are generally much more appropriate to measure fluids or (semi-)transparent samples. In this context one should mention a recent paper by Gendre et al. [7]. The authors presented a method to measure and calculate thermal diffusivity by the modulation technique which originated from photothermal radiometry. Some mathematics exposed in their article and in the present article are partly comparable. Their paper can be considered to form a primary link between the "classical" and the "alternative" methods.

The incentive for the present reanalysis of Cowan's theoretical treatment is to simplify further the mathematics for the general case. This may lead to a wider use of the method. Also, a discussion is given of possible inaccuracies and some suggestions for further improvements.

The basic features exposed in this paper have been reported earlier in a conference, but only an abstract of the text was published [8].

2. REANALYSIS OF COWAN'S THEORETICAL TREATMENT USING COMPLEX NUMBERS MATHEMATICS

It is assumed that one surface of a semiinfinite sample (see Fig. 1) is uniformly heated by a continuous energy beam with a small sinusoidal modulation superimposed on it. Also, it is assumed that energy conduction takes place through the sample perpendicular to the surface only and that loss of energy occurs only by radiation at both surfaces. Loss of energy by radiation only implicates vacuum conditions, e.g., in the case of electron beam heating, and thus no convection occurring. These assumptions include linearization of the energy transport and of the temperature distribution throughout the sample.

The total energy per unit surface reaching the surface at $x=l$ at time t is

$$Q(t)=Q_{\mathrm{c}}+Q_{\mathrm{m}}(t) \tag{2}$$

being the sum of a constant part Q_{c} and of a sinusoidally modulated part $Q_{\mathrm{m}}(t)$.

The temperature distribution at any plane x throughout the sample at time t can thus be represented by

$$T(x, t)=T_{\mathrm{c}}(x)+T_{\mathrm{m}}(x, t) \tag{3}$$

Linearization implies, among other things, that the temperature distribution throughout the sample is governed by the one-dimensional differential heat diffusion equation:

$$a \frac{\partial^2 T(x, t)}{\partial x^2}=\frac{\partial T(x, t)}{\partial t} \tag{4}$$

where a is the thermal diffusivity, which is by definition equal to

$$a=\frac{\lambda}{\rho C_{\mathrm{p}}} \tag{5}$$

with λ thermal conductivity, ρ density, and C_{p} specific heat.

In order to solve Eq. (4) for this particular situation it is first split into two distinct parts in accordance with Eq. (3), the first part handling the constant (or equilibrium) terms

$$a \frac{\partial^2 T_{\mathrm{c}}(x)}{\partial x^2}=0 \tag{6}$$

and the second part the modulation term, which is of direct interest here:

$$a \frac{\partial^2 T_{\mathrm{m}}(x, t)}{\partial x^2}=\frac{\partial T_{\mathrm{m}}(x, t)}{\partial t} \tag{7}$$

In this stage of the analysis all reasoning exposed is still essentially identical to that of Cowan. Yet when treating time-dependent phenomena and associated phase shifts, it is advantageous to make use of complex numbers mathematics.

In the linearized problem the energy modulation and the temperature are sinusoidal functions of time. Thus

$$Q_{\mathrm{m}}(t)=q e^{i \omega t} \tag{8}$$

(where $\omega = 2\pi f$ is the angular frequency) and

$$T_m(x, t) = \theta(x)\, e^{i\omega t} \tag{9}$$

In Eqs. (8) and (9), q and $\theta(x)$ are complex quantities.

As a result, the heat diffusion equation (7), dealing with the modulation part, becomes

$$a\left[\frac{d^2\theta(x)}{dx^2}\right] = i\omega\theta(x) \tag{10}$$

By introducing the parameter

$$\beta = \left(\frac{i\omega}{a}\right)^{1/2} = (1+i)\left(\frac{\omega}{2a}\right)^{1/2} \tag{11}$$

(which may be called the thermal diffusion length), Eq. (10) can be written as

$$\frac{d^2\theta(x)}{dx^2} = \beta^2\theta(x) \tag{12}$$

and the solution is of the form

$$\theta(x) = A_+ e^{\beta x} + A_- e^{-\beta x} \tag{13}$$

where the constants A_+ and A_- are complex numbers.

In order to determine these constants, appropriate boundary conditions have to be introduced.

A first boundary condition can be established since the energy balance at the nonheated face, where $x = 0$, is the result of heating by conduction through the sample and of cooling by the radiative energy loss from that surface:

$$\lambda_0 \frac{\partial T(0, t)}{\partial x} = \varepsilon(0, t)\, \sigma T^4(0, t) \tag{14}$$

where $\varepsilon(0, t)$ is the total thermal emissivity at $x = 0$ and at time t, σ is the Stefan–Boltzmann constant, and $T(0, t)$ is the temperature at $x = 0$ and at time t.

Developing Eq. (14), taking Eq. (3) into account, leads to

$$\lambda_0 \frac{\partial T_c(0)}{\partial x} + \lambda_0 \frac{\partial T_m(0, t)}{\partial x} = \varepsilon(0, t)\, \sigma[T_c(0) + T_m(0, t)]^4 \tag{15}$$

The right-hand term can be expanded following a Taylor series whereby the higher-order terms can be neglected since the modulation depth is assumed to be small. Consequently in the neighbourhood of $T_c(0)$ one can write

$$\lambda_0 \frac{\partial T_c(0)}{\partial x} + \lambda_0 \frac{\partial T_m(0, t)}{\partial x} = \varepsilon(0, t)\, \sigma T_c^4(0) + T_m(0, t) \frac{\partial[\varepsilon(0, t)\, \sigma T_c^4(0)]}{\partial T} \tag{16}$$

This equation can be split into two equations:

$$\lambda_0 \frac{\partial T_c(0)}{\partial x} = \varepsilon(0, t)\, \sigma T_c^4(0) \tag{17}$$

$$\lambda_0 \frac{\partial T_m(0, t)}{\partial x} = T_m(0, t) \frac{\partial[\varepsilon(0, t)\, \sigma T_c^4(0)]}{\partial T} \tag{18}$$

By introducing the notation

$$c_0 = \frac{1}{\lambda_0} \frac{d[\varepsilon(0, t)\, \sigma T_c^4(0)]}{dT} = \frac{1}{\lambda} \frac{d[\varepsilon \sigma T_c^4(0)]}{dT} \cong \frac{1}{\lambda} 4\varepsilon\sigma T_c^3 \tag{19}$$

(c_0 may be called the inverse extrapolation length), Eq. (18) can be written as follows, also accepting $\varepsilon(0, t) = \varepsilon$ and $\lambda_0 = \lambda$, which means that the emissivity and the conductivity remain constant with time and space, thus also within the small modulated temperature range:

$$\frac{dT_m(0, t)}{dx} = T_m(0, t)\, c_0 \tag{20}$$

Equation (20) can further be transformed, by taking Eq. (9) into account, to

$$\frac{d\theta(0)}{dx} = \theta(0)\, c_0 \tag{21}$$

Now the solution of Eq. (13) can be worked out. At $x = 0$ it may be concluded from Eq. (13) that

$$\theta(0) = A_+ + A_- \tag{22}$$

and that

$$\frac{d\theta(0)}{dx} = \beta A_+ - \beta A_- \tag{23}$$

Combining Eqs. (21), (22), and (23) leads to

$$c_0(A_+ + A_-) = \beta(A_+ - A_-) \tag{24}$$

This allows one to determine A_+ and A_- up to a proportionality factor P, by transforming Eq. (24) to Eqs. (25) and (26), respectively:

$$A_+ = (\beta + c_0)\, P \tag{25}$$

$$A_- = (\beta - c_0)\, P \tag{26}$$

As only differences of phases are of importance, a suitable reference phase may be imposed for which the phase angle of the complex number is zero (the phase angle of P may be put equal to zero), while the modulus is irrelevant.

Equation (13), the complex function of $\theta(x)$, may now be computed at $x=0$ and at $x=l$, up to a real factor P, from which the phase angles Φ_0 and Φ_l can be calculated readily when taking into account Eqs. (11), (25), and (26):

$$\Phi_0 = \Phi[\theta(0)] = \Phi[A_+ + A_-] = \Phi[2\beta P] = \frac{\pi}{4} \tag{27}$$

$$\Phi_l = \Phi[\theta(1)] = \Phi[(\beta + c_0)\, e^{\beta l} + (\beta - c_0)\, e^{-\beta l}] \tag{28}$$

A second boundary condition is now introduced. At the heated face $x=l$, the energy flowing into the sample by conduction will be the difference between the energy deposited and the energy lost by radiation at that surface. Consequently,

$$\lambda_l \frac{\partial T(l,t)}{\partial x} = Q(t) - \varepsilon(l,t)\, \sigma T^4(l,t) \tag{29}$$

At that face, a reasoning analogous to that used before (the boundary condition at $x=0$) leads to the following two equations:

$$Q_c = \lambda_l \frac{\partial T_c(l)}{\partial x} + \varepsilon(l,t)\, \sigma T_c^4(l) \tag{30}$$

$$Q_m(t) = \lambda_l \frac{\partial T_m(l,t)}{\partial x} + T_m(l,t) \frac{\partial[\varepsilon(l,t)\, \sigma T_c^4(l)]}{\partial T} \tag{31}$$

Again, by introducing the notation

$$c_1 = \frac{1}{\lambda_1} \frac{d[\varepsilon(l,t)\, \sigma T_c^4(l)]}{dT} = \frac{1}{\lambda} \frac{d[\varepsilon\sigma T_c^4(l)]}{dT} \cong \frac{1}{\lambda} 4\varepsilon\sigma T_c^3(l) \tag{32}$$

Equation (31) can be written as follows, also again accepting $\varepsilon(l, t) = \varepsilon$ and $\lambda_l = \lambda$:

$$Q_{\mathrm{m}}(t) = \lambda \frac{dT_{\mathrm{m}}(l, t)}{dx} + T_{\mathrm{m}}(l, t)\, \lambda_l c_l \tag{33}$$

and after having entered Eqs. (8) and (9),

$$q = \lambda \frac{d\theta(l)}{dx} + \theta(l)\, \lambda c_l \tag{34}$$

For the adopted conventions the phase angle (Φ_q) of the energy beam with respect to the reference is now computed from Eq. (34) taking into account $x = l$ and Eqs. (13), (25), and (26):

$$\Phi_q = \Phi\left[\frac{q}{\lambda}\right] = \Phi[(\beta + c_0)(\beta + c_1)\, e^{\beta l} - (\beta - c_0)(\beta - c_1)\, e^{-\beta l}] \tag{35}$$

At this stage, one can compute the phase shifts between q, $\theta(0)$, and $\theta(l)$ in a straightforward manner by operations with complex numbers.

The two phase shifts, with reference to the energy beam, will be

$$\delta_0 = \Phi_q - \Phi_0 = \Phi_q - \frac{\pi}{4} \tag{36}$$

$$\delta_l = \Phi_q - \Phi_l \tag{37}$$

and consequently, since $\Delta = \delta_0 - \delta_1$ [Eq. (1)],

$$\Delta = \Phi_l - \frac{\pi}{4} \tag{38}$$

These equations do not need an explicit development.

3. ALGORITHM TO COMPUTE THE RESULTS

Equation (38), expressing Δ as a function of a number of parameters, enables in principle the calculation of the thermal conductivity λ or of the thermal diffusivity a. However, as the equations contain implicit functions of the conductivity (or of the diffusivity), the solution has to be derived by iteration.

The two measured quantities are the temperature and Δ. Since the temperature is measured by pyrometry, corrections are necessary for the transmission of the measuring windows and for either the spectral emissivity of the sample, when utilizing a brightness pyrometer, or the ratio of the spectral emissivities, when a two-color pyrometer is used. For these corrections see Ref. 9. First, the parameters thickness l, thermal expansion coefficient α, density ρ, specific heat C_p, and total emissivity ε are introduced (either measured values or taken from literature) and their effective value is calculated for the temperature considered. Then, after selection of a starting value for λ [or for a; see Eq. (5)], the corresponding value for $\varDelta$ is calculated through Eq. (38), taking into account Eqs. (28), (11), and (19). This calculated $\varDelta$ is compared with the measured $\varDelta$. Iteration goes on each time using a new adjusted value for λ (or a), until the calculated and the measured values of $\varDelta$ are in agreement. At that moment the final value for λ (or a) has been obtained. During each iteration step, the temperature will be adjusted slightly and all the parameters involved have to be adjusted accordingly.

4. DISCUSSION AND CONCLUSIONS

It has been shown that the use of complex numbers notation leads to rather simple expressions for $\varDelta$. Indeed, the development exposed here contains nothing else but sums and ratios of the exponential functions of complex numbers.

A direct advantage of the complex numbers method is that the present algorithm covers the range of phase lags from 0 to 360°, whereas in the Cowan or the Penninckx method a phase lag of 180° cannot be exceeded since δ_l, δ_0, and $\varDelta$ are expressed by a tangent. This makes the method more universal, and moreover, the troubles possibly experienced with a tangent around 90° are now avoided.

A comparative computation has been carried out of the results obtained with the method proposed here and with the methods of respectively Cowan [1, 2], Wheeler [4], Penninckx [6], Schmidt [10], and Brandt and Neuer [11], each time using identical fictive test data. Exact agreement was found for all methods, although completely different algorithms were involved each time.

Next to this comparative examination, also an estimate of the absolute accuracy should be considered. It has to be mentioned that the present evaluation is adapted to our method and equipment (see Ref. 12), although in principle it may apply equally well to other methods used for measuring thermal diffusivity.

Apart from possible measurement errors (phase, temperature, etc.) and from errors arising from experimental conditions (e.g., nonisothermal heating, etc.), the final equations contain some theoretical simplifications, mainly as a result of the mathematical linearization. It is not obvious whether these simplifications always introduce negligible errors in practice. They may be reviewed as follows.

1. *Requirement for a semiinfinite sample, thermal conduction perpendicular to the sample surface only and loss of energy by radiation only.* These requirements can be met sufficiently well by choosing adequate dimensions for the sample (∅ 8 mm, ≅1 mm thick) and for the sample holder with guard ring. Semiinfinity is, moreover, strongly enhanced when only the central part of the specimen is measured (e.g., 1/10 of the upper and lower surfaces) where perpendicular conduction prevails. Loss by conduction through the sample holder suspending wires can be kept negligible. The surface in contact is only some 0.1% of the total surface or even less. Loss by convection is not occurring since heating by an electron beam requires an evacuated environment anyhow.
2. *Linearization of the energy transport through the sample.* Even for very poor thermal conductors the difference between the mean temperatures of the bombarded and of the nonbombarded surface will normally be less than 1 to 2% of the mean temperature and the modulation depth can generally be kept below some 0.5%. Over such a small temperature range, linear transport can be assumed.
3. *Substitution of* $\varepsilon(l, t) = \varepsilon(0, t)$ *by* ε *and* $\lambda_l = \lambda_0$ *by* λ *in Eqs.* (19) *and* (32). This means that the total emissivity as well as the thermal conductivity is supposed to be constant within the temperature interval over the sample. This condition is almost always fulfilled, in view again of the very small temperature differences, except possibly in close proximity of a concurrent phase transition.
4. *Boundary conditions.* The two boundary conditions, dealing with the energy balance between conduction and radiative heat loss [Eqs. (14) and (29)] are valid for radiation loss into an infinite space at 0 K. Yet since radiation will occur toward the measuring chamber walls, which are usually almost at room temperature, an error of a few percent is possible when the specimen temperature is, e.g., 500°C or less. Reduction of this error is easily achieved by taking into account the actual wall temperature through the $(T_c^4 - T_{\mathrm{wall}}^4)$ radiative heat loss relation. At higher temperatures the error will always be negligible.

An inherent weakness of all thermal diffusivity methods is the fact that the various interfering parameters are not always known with a sufficiently high accuracy. The influence of these parameters on the final results for the

Table I. Average Deviation of the Final Calculated Results (T, a, and λ) as Caused by an Error Value of 10% for the Various Interfering Parameters

Interfering parameter, differing from real value by 10%	Deviation of final calculated results (%)			Final significance
	T	a	λ	
T	11	0.4	6	left out of consideration[a]
Δ	0.05	13	13	
f	0.005	1	1	Can be neglected
l	0.03	19	19	Important for a and λ
α	0.005	0.3	0.2	Negligible
ε	0.03	0.2	0.2	Negligible
ε_s	3	0.1	2	Low to negligible
ρ	0.03	0.2	10	Important for λ
C_p	0.03	0.2	10	

[a] Mentioned only for completeness since T and Δ do not directly interfere in this study.

diffusivity a, the conductivity λ, and the temperature T has been examined numerically. Table I gives the average deviations of the finally calculated results as caused by a deviation of 10% for each interfering parameter.

T and Δ are measured data. Their accuracy depends primarily on the instruments used. The value of f is normally known very accurately. An error on the parameter l has a large influence both on the diffusivity and on the conductivity, whereas the values of C_p and of ρ influence only the conductivity. An error in the parameters α, ε and ε_s leads only to a negligible or small deviation of the calculated results (although it should be emphasized that ε and ε_s are very often hardly known).

The inherent weakness in connection with the various imperfectly known interfering parameters can possibly be obviated by determining one of these parameters, simultaneously with T, a, and λ. Therefore, a second measurement is necessary, e.g., δ_l or δ_0, next to Δ. In a forthcoming paper this possibility will be treated more extensively. It should be mentioned that Schmidt [10], Brandt and Neuer [11], and Zhou Ben-Lian [13] have made progress in this direction.

NOMENCLATURE

a	Thermal diffusivity, $m^2 \cdot s^{-1}$
c	Index denoting a constant part, dimensionless
c_l, c_0	Inverse extrapolation length, m^{-1}

C_p	Specific heat, $\mathrm{J \cdot kg^{-1} \cdot K^{-1}}$
f	Modulation frequency, Hz
l	Thickness of disk-shaped sample, m
Q_c	Equilibrium energy per unit surface deposited on surface $x = l$, $\mathrm{W \cdot m^{-2}}$
$Q_\mathrm{m}(t)$	Energy of modulation per unit surface deposited on surface $x = l$, $\mathrm{W \cdot m^{-2}}$
$Q(t)$	Total energy per unit surface deposited on surface $x = l$, $\mathrm{W \cdot m^{-2}}$
q	Complex energy modulation amplitude, $\mathrm{W \cdot m^{-2}}$
T_l	Equilibrium temperature of heated surface, K
T_0	Equilibrium temperature of nonheated surface, K
$T(x, t)$	Total temperature of any plane at distance x and at time t, K
$T_\mathrm{m}(x, t)$	Modulation temperature at any distance x and at time t, K
t	Time, s
x	Distance perpendicular to the specimen's surface and with the nonheated surface as the reference, m
α	Thermal linear expansion coefficient, dimensionless
β	Intermediary parameter, $\mathrm{m^{-2}}$
Δ	Phase difference between heated and nonheated specimen face, radian
δ_0	Phase difference between energy modulation and nonheated face, radian
δ_l	Phase difference between energy modulation and heated face, radian
ε	Total emissivity, dimensionless
ε_s	Spectral emissivity, dimensionless
θ	Temperature, amplitude of modulated part argument, K
λ	Thermal conductivity, $\mathrm{W \cdot m^{-1} \cdot K^{-1}}$
ρ	Density, $\mathrm{kg \cdot m^{-3}}$
σ	Stefan–Boltzmann constant, $5.66961 \times 10^{-8} \mathrm{W \cdot m^{-2} \cdot K^{-4}}$
ω	Angular frequency $= 2\pi f$, $\mathrm{s^{-1}}$

Indices used, (x), (x, t), c, m, l, 0, t, or the like, or combinations of them, denote a situation at x, or x and t, etc.

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