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Revisiting transient heat transfer in coated transparent media

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Abstract

Measurements of transparent materials are required to assess their conductive properties for industrial applications. This is typically done with a laser flash analysis (LFA) instrument, where a transient temperature response is analysed. Due to the multi-modality of heat transfer, the conductive contribution must be separated from e.g. the radiative transfer. Depending on the required complexity, this can be done either with a multi-parametric model or, in the diathermic case, with a model based on just one extra parameter, $\eta$. Unfortunately, this parameter has not been quantified based on any valid physical description. Furthermore, the simplified model solved analytically occasionally produced unexpected results, suspected to originate from the specific implementation. A correct treatment for the diathermic model is given in this paper, both in the analytical and numerical cases. Tests with polycrystalline alumina (PCA) and sapphire on two LFA setups were made to delineate the limits of applicability of the diathermic model. Finally, a revised version is proposed for both the diathermic and the coupled model to include convective losses and rear-face heating due to flashthroughs or circumferential heat fluxes.

Keywords: laser flash analysis, transparent materials, radiative transfer, coating, stray light, convection
1 Introduction

There is an interest in accurately measuring the thermal diffusivity of transparent materials. Several approaches have been developed to achieve this with the laser flash analysis (LFA, also simply known as the flash method) e.g. [1–7]. The experimental scheme usually involves coating the sample with graphite or a reflective coating on top of which graphite is sprayed. Either leads to the flash being absorbed at the boundaries, meaning only thermal radiation has to be accounted for in the model. A numerical solution to the coupled radiative-conductive problem describing heat transfer in a participating (emitting, absorbing and scattering) medium has recently been proposed in [7]. Unfortunately, the data collected with an area-averaging infrared detector of an LFA instrument is often not sufficient to ensure smooth operation of standard optimisers used in conjunction with the coupled model to analyse the time-temperature profiles measured experimentally [2, 7]. Therefore, it may seem more pragmatic to use a simpler treatment originally proposed in [3, 8]. The latter is built upon a standard model for an opaque material by Cowan [9], introducing only one extra parameter, the diathermic factor $\eta$, to treat thermal radiation. Compared to the coupled system of equations, this reduces the number of radiative transfer parameters from 3+ (optical thickness, refractive index, scattering albedo and the phase function parameters) to just one. In this case, model parameters are almost guaranteed to be non-collinear during optimisation. A naïve derivation of the system of equations by Mehling et al. [3] is available in [7] based on the radiative flux expression for two parallel grey surfaces. As it was not derived explicitly from a more general case, the applicability of Mehling’s model has largely remained an open question. And yet, the latter became popular after having been implemented in commercial software for LFA instruments [10]. There are no benchmarks available, which could unambiguously show whether the solution or its implementation are valid. However, there are reports some implementations of Mehling’s model fail [11, 12]. This is unsurprising, considering it took many years [13, 14] to come up with a correct analytical solution of the standard model for opaque samples proposed initially by Cape and Lehman [15]. Therefore, a careful revision is needed for the diathermic treatment proposed originally in [3, 8]. In addition to this mathematical aspect, there are some concerns on the validity of the problem statement, which required Brunner and Thermitus [10] to consider modified equations. In this connection, it is important to mention recent observations [16] of substantial convective heat losses in the standard LFA setups. Neither the diathermic nor the coupled model seem to explicitly take convective losses into account. Finally, the coating quality might also influence the results, as initially proposed in [8] and confirmed recently in [7]. With regards to the above, this paper aims to elucidate three aspects related to transient heat transfer in LFA experiments with transparent coated samples. Firstly, the applicability of the simplified diathermic model is explored. Secondly, the most effective approaches in solving the original system of equations are highlighted.
Ultimately, a modification to both the diathermic and coupled models is considered, allowing to extend its applicability to setups with a strong influence of convection or stray light.

2 Simplifying a coupled system

A set of equations describing one-dimensional heat transfer in a conductive-radiative environment within a grey enclosure was proposed in [7]:

\[
\frac{\partial \theta}{\partial \text{Fo}} = \frac{\partial^2 \theta}{\partial y^2} + \frac{\tau_0}{N_P} \times \left( -\frac{dq}{d\tau} \right), \quad 0 < y < 1, \quad \text{Fo} > 0, \quad (1a)
\]

\[
\left. \frac{\partial \theta}{\partial y} \right|_{y=0} = \text{Bi}(r) \cdot \theta_{y=0} + \frac{1}{N_P} q(0) - \Phi(\text{Fo}), \quad (1b)
\]

\[
\left. \frac{\partial \theta}{\partial y} \right|_{y=1} = -\text{Bi}(r) \cdot \theta_{y=1} + \frac{1}{N_P} q(1), \quad (1c)
\]

\[
\theta(\text{Fo} = 0) = 0, \quad (1d)
\]

where \( q(y) \) is the radiative flux computed by solving the radiative transfer equation (RTE); \( N_P = \lambda/(4\sigma_0 n^2 T_0^3 l) \) is the Planck number; \( n \) is the refractive index; \( \tau_0 \) and \( \tau \) are the optical thickness and the optical coordinate respectively. \( \text{Bi}(r) = 4\sigma_0 \varepsilon T_0^3 l/\lambda \) is the radiative Biot number; \( \theta = (T - T_0) \cdot C_p \rho \pi d^2 l/(4Q) \) is the dimensionless heating; \( \text{Fo} = at/l^2 \) is the Fourier number; \( \lambda \) and \( a \) are the thermal conductivity and diffusivity respectively; \( \varepsilon \) is the emissivity; \( l \) and \( d \) are sample thickness and diameter; \( y = x/l \) is the dimensionless coordinate; \( T_0 \) is the test temperature. Additionally, \( \Phi(\text{Fo}) \) is the pulse function measured with the pulse diode.

It is argued that a much simpler set of equations may be derived under certain simplifying conditions. These simplifications are addressed in the following subsections.

2.1 Optically thin, non-scattering case

In a non-scattering medium the radiative heat transfer equation is vastly simplified. Assuming diffuse emission and reflection at the boundaries, the boundary fluxes may be calculated as [7]:

\[
q(0)/\pi = i^+(0) - 2i^-(\tau_0)E_3(\tau_0) - 2 \int_0^{\tau_0} j(t)E_2(t)dt,
\]

\[
q(1)/\pi = -i^-(\tau_0) + 2i^+(0)E_3(\tau_0) + 2 \int_0^{\tau_0} j(t)E_2(\tau_0 - t)dt.
\]

where \( j(t) = 1/(4\pi)T_0/\delta T_m [1 + \theta(t)\delta T_m/T_0]^4 \) and \( E_n(x) \) is the \( n \)-th order exponential integral.
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In a optically thin medium τ₀ ≪ 1. Consequently, E₂(τ₀) ≃ 1 and E₃(τ₀) ≃ 1/2. Moreover, it can be shown [17] that dq/dτ ≡ 0. The second term in (1a) may thus be omitted. Furthermore, boundary intensities are simply:

\[ i^+(0) = \frac{j(0) + j(\tau_0)(1 - \varepsilon)}{2 - \varepsilon}, \]
\[ i^-(\tau_0) = \frac{j(\tau_0) + j(0)(1 - \varepsilon)}{2 - \varepsilon}. \]

Finally, when δTᵣ ≪ T₀, i.e., the flash power is low:

\[ q(0) \equiv q(1) = \frac{\varepsilon}{2 - \varepsilon} (\theta(0) - \theta(\tau_0)). \] (2)

The right-hand side in (2) may also be derived from elementary considerations e.g. taking the radiative flux between two infinite parallel planes in thermal equilibrium cf. [7], or in the two-flux approximation for a diathermic material cf. Section 2.2.

The diathermic set of equations may then be derived from the coupled problem statement by substituting (2) into (1b), (1c) and noticing the second term in the right hand side in (1a) vanishes. This gives:

\[ \frac{\partial \theta}{\partial F₀} = \frac{\partial^2 \theta}{\partial y^2}, \quad 0 < y < 1, \quad F₀ > 0, \] (3a)
\[ \left. \frac{\partial \theta}{\partial y} \right|_{y=0} = B_{i(r)} \cdot \theta_{y=0} + \eta B_{i(r)} (\theta_{y=0} - \theta_{y=1}) - \Phi(F₀), \] (3b)
\[ \left. \frac{\partial \theta}{\partial y} \right|_{y=1} = -B_{i(r)} \cdot \theta_{y=1} + \eta B_{i(r)} (\theta_{y=0} - \theta_{y=1}), \] (3c)
\[ \theta(F₀ = 0) = 0, \] (3d)

where the diathermic factor

\[ \eta = n^2/(2 - \varepsilon) \] (4)

is introduced.

The condition \( \eta \leq 1 \) previously imposed in [7] is only valid when \( n = 1 \), which corresponds to vacuum. Perhaps due to a typo, the numerator of \( \eta \) in [7] was given incorrectly as \( \varepsilon \). The correct expression (4) indicates that, under favourable conditions, \( \eta \) may reach values greater than unity. Furthermore, in the non-scattering case: \( \eta > 1/2 \).

2.2 Isotropically scattering medium

Consider an idealised non-absorbing material, which scatters radiation isotropically. The scattering coefficient is denoted as \( \chi \). A two-stream model is used
to derive an expression for $\eta$ in this case. The positive stream gives rise to the flux $q_1$:

$$\frac{\partial q_1}{\partial y} = -\chi q_1 + \chi q_2.$$ 

By analogy, the negative stream defines $q_2$:

$$\frac{\partial q_2}{\partial (-y)} = -\chi q_2 + \chi q_1.$$ 

Therefore $q_1 - q_2 = J$, where $J$ is an unknown quantity equal to the energy flux transferred between the sample faces. Let $q_{k,1}$ and $q_{k,2}$ be the values of the fluxes at the front and rear faces correspondingly. It follows from the above that:

$$q_{k,2} = q_{k,1} - \chi Jl.$$ 

(5)

The flux balance equations for the front and rear faces are:

$$q_{1,1} = I_1 + (1 - \varepsilon)q_{2,1},$$

(6a)

$$q_{2,2} = I_2 + (1 - \varepsilon)q_{1,2},$$

(6b)

where $I_i = n^2 \sigma \varepsilon T_i^4$, $i = 1, 2$ is the integrated spectral radiance given by the Stefan-Boltzmann law, $T_i$ is the temperature at either $y = 0$ or $y = 1$.

Substituting (5) into (6b) gives:

$$q_{2,1} - aJL = I_2 + (1 - \varepsilon)(q_{1,1} - \chi Jl).$$

(7)

Subtracting (7) from (6a) gives:

$$J = (I_1 - I_2)/(2 - \varepsilon + \varepsilon \chi l).$$

Finally:

$$\eta = \frac{n^2}{2 - \varepsilon + \varepsilon \chi l}.$$ 

(8)

This result shows that $\eta$ may reach values well below $1/2$ in samples with isotropic scattering.

### 2.3 Optically thick limit

Hahn et al. [8] defined the coefficient $\eta$ for an optically thick medium in the so-called Rosseland approximation as $n^2/[1 + 4/3\tau_0]$, with reference to [18]. It was shown in [17] that the fluxes calculated within the Rosseland approximation are only strict at large $\tau$ and $\tau_0 - \tau$ values. This means getting incorrect values for the boundary fluxes $q(0)$ and $q(1)$. Therefore, (3a) – (3d) is inapplicable in the optically thick limit.
3 Solving the simplified system

3.1 Analytical solution

Hahn et al. [8] had previously shown that the analytical solution \( \theta(y, \text{Fo}) \) to (3a) – (3d) with \( \Phi(\text{Fo}) = \delta(\text{Fo}) \) is similar to the one given by Cowan [9] for a one-dimensional thermal transient in a fully opaque solid cylinder where a special form of the transcendental equation is used. An alternative derivation of this solution for an arbitrary pulse shape is given in this section.

Instead of having the heat source \( \Phi(\text{Fo}) \) in (3b), the latter may be included in (3a), see [8]:

\[
\frac{\partial \theta}{\partial \text{Fo}} = \frac{\partial^2 \theta}{\partial y^2} + \Phi(\text{Fo}) \delta(y),
\]

where \( \delta(y) \) is the Dirac delta function for the pulse at \( y = 0 \).

The boundary conditions are then:

\[
\left. \frac{\partial \theta}{\partial y} \right|_{y=0} = \text{Bi}(r) \cdot \theta_{y=0} + \eta \text{Bi}(r) (\theta_{y=0} - \theta_{y=1}), \quad (10a)
\]

\[
\left. \frac{\partial \theta}{\partial y} \right|_{y=1} = -\text{Bi}(r) \cdot \theta_{y=1} + \eta \text{Bi}(r) (\theta_{y=0} - \theta_{y=1}) \quad (10b)
\]

Note (10a) and (10b) are symmetric with respect to \( y' = y - 1/2 \). If \( \theta(y') \) is even, a substitution \( y \leftrightarrow y' \) in (10a) transforms it into (10b). This property also holds when \( \theta(y') \) is odd. Therefore, the eigenfunctions of the system formed by (9), (10a) and (10b) should satisfy:

\[
u_k = \cos(\lambda_k [y - 1/2]) \quad \text{(even modes)}, \quad (11a)\\
v_k = \sin(\mu_k [y - 1/2]) \quad \text{(odd modes)}, \quad (11b)
\]

Substituting (11a) – (11b) into either (10a) or (10b) leads to the following relations for even and odd modes:

\[
\lambda_k \tan(\lambda_k/2) = \text{Bi}, \quad (12a)\\
\lambda_k > 0, \quad k = 0, 1, 2, \ldots \quad \text{(even modes)}\\
\mu_k \cot(\mu_k/2) = -\text{Bi}' \quad (12b)\\
\mu_k > 0, \quad k = 0, 1, 2, \ldots \quad \text{Bi}' = \text{Bi}(1 + 2\eta) \quad \text{(odd modes)}
\]

To facilitate further analysis, the above eigenfunctions and eigenvalues can be grouped as:
\[ f_n = \begin{cases} u_k, & n = 2k \\ v_k, & n = 2k + 1 \end{cases}, \quad k = 0, 1, 2, \ldots \quad (13a) \]

\[ \beta_n = \begin{cases} \lambda_k, & n = 2k \\ \mu_k, & n = 2k + 1 \end{cases}, \quad k = 0, 1, 2, \ldots \quad (13b) \]

\[ B_i_n = \begin{cases} B_i, & n = 2k \\ B'_i, & n = 2k + 1 \end{cases}, \quad k = 0, 1, 2, \ldots \quad (13c) \]

The solution to (3a) – (3d) at \( y = 1 \) is then given by the convolution:

\[ \theta(F_o, 1) = \Phi(F_o) \ast \left( \sum_{n=0}^{\infty} \frac{f_n(0)f_n(1)}{(f_n \cdot f_n)} \exp(-\beta^2_n F_o) \right), \quad (14) \]

After substituting \( f_n \), this simplifies to [8]:

\[ \theta(F_o, 1) = \Phi(F_o) \ast \left( \sum_{n=0}^{\infty} \frac{(-1)^n 2\beta^2_n}{\beta^2_n + B_i^2_n + 2B_i n} \exp(-\beta^2_n F_o) \right). \quad (15) \]

Calculation uses the \( \beta_n \) roots to the transcendental equations (12a) and (12b), which must be found separately.

### 3.2.1 Implementation

Consider the transcendental equation (12a). By varying \( \lambda_k/2 \) from \( 2\pi k \) to \( 2\pi k + \pi \), a monotonic change of \( \tan(\lambda_k/2) \) from zero to \( +\infty \) is observed. At the same time, \( (\lambda_k/2)^{-1} B_i \) decreases monotonically. Therefore, (12a) has exactly one positive root, \( \lambda_k \), on each \([2\pi k, 2\pi k + \pi]\) interval. By analogy, (12b) has exactly one positive root, \( \mu_k \), on each \([2\pi k + \pi, 2\pi k + 2\pi]\) interval.

It is practical to search for the \( k \)-th root of (12a) and (12b) on the intervals \([2\pi k, 2\pi k + \pi - \varepsilon]\) and \([2\pi k + \pi, 2\pi k + 2\pi - \varepsilon]\) correspondingly, where \( \varepsilon \) is a machine epsilon. The most reliable method for root-finding within these intervals is the ZEROIN method by Brent and Dekker [19] combining the bisection method and inverse quadratic interpolation. An application of this method to finding the roots of (12a) and (12b) is available in the GNU Octave/Matlab function provided in the Supplementary Material.

A popular implementation of the diathermic model is based on using an alternative expression to (12a) and (12b) given in [3]

\[ \tan B_i_n = \frac{2\beta_n B_i_n}{\beta^2_n - B_i^2_n}, \quad \beta_n > 0, \quad n = 0, 1, 2, \ldots \quad (16) \]

and an alternative expression for (15) where the numerator \((-1)^n 2\beta^2_n\) is replaced by \( \beta_n[\beta_n \cos(\beta_n) + B_i n \sin(\beta_n)] \).
We argue that using (12a), (12b) and (15) has a certain advantage over this latter approach. Firstly, the right-hand side of (16) has an extra pole at \( \beta_n = \text{Bi}_n \), which complicates the interval selection for the root finder. Secondly, the numerator with trigonometric functions, although equivalent to \((-1)^n \beta_n^2\) mathematically, is computationally less accurate.

Another point of concern is the root-finding routine. Most applications rely on the unconstrained Newton’s method. Under less favourable conditions, large increments may lead to the Newton’s method generating a \( k \)-th approximation of the root, which lies outside of the selected interval. This may result in a neighbouring root being found and wrongly assigned to the \( k \)-th root, leading to an erroneous set of roots \( \beta_n \).

Methods such as bisection or ZEROIN [19] generate roots strictly confined within the given intervals and are recommended for use.

### 3.3 Numerical solution

#### 3.3.1 Difference scheme

Calculating the convolution in (15) is possible when certain assumptions are made on the shape of the pulse function \( \Phi(F_0) \), see [14]. Unfortunately, the pulse diode data is not always guaranteed to fit a selected analytical expression. Moreover, numerical instabilities may arise when the convolution is calculated at large pulse widths. Other complications include selecting the number of terms in (15), which should be sufficiently large to ensure convergence at any given \( F_0 \) value.

Ironically, to calculate \( \theta(F_0, 1) \) derived analytically, one must rely on numerical methods. What if a purely numerical procedure was chosen from the beginning? A finite-difference solution in [7] fulfils this goal.

Consider a four-point implicit difference scheme in Figure 1 for (3a) – (3d), which is constructed on a uniform grid and a constant time step \( \tau \). Let the superscript \( i \) and the subscript \( j = 0, \ldots, N - 1 \) denote the time index and the coordinate index respectively. The following relation holds between the time step and the grid spacing \( h \): \( \tau = \tau_f h^2 \), where \( \tau_f \leq 0.5 \). The layer at time index \( i + 1 \) contains unknown functions values. Using this scheme to discretize both the heat equation (3a) and the boundary conditions (3b) and (3c) leads to the following equivalent matrix equation:

\[
A \theta^{i+1} = R, \tag{17a}
\]

\[
A = \begin{pmatrix}
  z_0 & -1 & 0 & 0 & 0 & \cdots & 0 & 0 & z_{N-1} \\
  1 & -b_1 & 1 & 0 & 0 & \cdots & 0 & 0 & 0 \\
  0 & 1 & -b_2 & 1 & 0 & \cdots & 0 & 0 & 0 \\
  \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots \\
  0 & 0 & 0 & 0 & 0 & \cdots & 1 & -b_{N-2} & 1 \\
  z_{N-1} & 0 & 0 & 0 & 0 & \cdots & 0 & -1 & z_0
\end{pmatrix}, \tag{17b}
\]
The four-point difference scheme approximating the heat equation (3a) and the boundary conditions (3b) and (3c), where the heating function $\theta_i$, $i = 0, 1, 2, ..., j = 0, 1, ..., N - 1$. A uniform grid with spacing $h$ is introduced. A constant time step is equal to $\tau$.

$$R = (r_0 \ R_1 \ R_2 \ \cdots \ R_{N-2} \ r_{N-1})$$

(17c)

where $b_j = 2 + h^2/\tau; R_j = -h^2/\tau^j_i; z_0 = 1 + h^2/2\tau + hBi(1+\eta); z_{N-1} = -h\eta Bi; r_0 = h^2/2\tau^0_i + h\Phi^i+1$ and $r_{N-1} = h^2/2\tau^{N-1}i$.

In opaque materials ($\eta = 0$), the matrix $A$ is tridiagonal, which allows using the Thomas algorithm to solve the linear set. However, this is not the case at $\eta > 0$. The matrix $A$ in (17b) is composed of an inner tridiagonal block and a border formed by an extra row and column. This block system is solved using the Sherman–Morrison–Woodbury identity and the Thomas algorithm for the main block, as previously discussed in [7]. This results in closed-form expressions for the heating $\theta^i+1_j$ at $j = 0, ..., N - 1$:

$$\theta_j = w^I_j + \theta^I_{N-1} w^{II}_j, \ j = 0, ..., N - 2,$$

$$\theta^I_{N-1} = (r_{N-1} - z_{N-1} w^I_0 + w^{II}_{N-2})/(z_0 + z_{N-1} w^{II}_0 - w^{II}_{N-2}),$$

$$(18a)$$

$$(18b)$$

$$w^{I}_j = \alpha_{j+1} w^{I}_{j+1} + \beta_{j+1}, \ j = N - 3, ..., 0,$$

$$w^{II}_{N-2} = \beta_{N-1}, \ w^{II}_{N-2} = \alpha_{N-1} + \gamma_{N-1},$$

$$\alpha_1 = 1/z_0, \ \beta_1 = r_0/z_0, \ \gamma_1 = -z_{N-1}/z_0, \ \alpha_{j+1} = c_j/(b_j - a_j \alpha_j), \ \beta_{j+1} = (R_j - a_j \beta_j)/(a_j \alpha_j - b_j),$$

$$\gamma_{j+1} = a_j \gamma_j / (b_j - a_j \alpha_j), \ j = 1, ..., N - 2.$$  

(18c)

This solution is $O(h^2 + \tau)$ accurate.

### 3.3.2 Implementation

As discussed in [7], it is sufficient to choose a grid with $N = 30$ points, which corresponds to $h = 1/30$. The time step should ideally be selected based on the pulse width, which is always finite in LFA measurements. However, it is not practical to consider pulses shorter than, e.g., $1/10,000$ of the characteristic time $l^2/a$. This requires a very small time step and does not result in an
appreciable rise in accuracy. Finite-pulse effects are usually only pronounced when the pulse width is larger than $1/100$ of the half-time $t_{1/2}$. Therefore, the minimum time step is selected as $10^{-4}l^2/a$. At large pulse widths, the discretisation of the pulse shape starts to significantly affect the shape of the heating curve. According to (18a) – (18c), the pulse function $\Phi(F_o)$ is calculated at discrete time steps $i\tau, (i+1)\tau, \ldots$. To ensure that the maximum heating $T_{\text{inf}} = (C_p\rho\pi d^2l)/4Q$ and the shape of the heating curves are calculated correctly, the discrete representation of the pulse function $\Phi^i$ should capture most information of the true pulse diode shape $\Phi(F_o)$. This is required when the variations in the pulse power $\Phi$ occur at the time scale of the variations in $\theta$.

To achieve this, $\Phi^i$ should be calculated at a sufficiently large number of time steps. Practice shows that a total of 20 points is sufficient in most cases. This means having a time step at least 20 times shorter than the pulse width. Finally, $\Phi^i$ should be normalised to give $P^i_0(\Phi^i + \Phi^{i+1})\tau = 1$. This is simply done by scaling the values of the pulse diode data uniformly. At intermediate pulse widths, the level of detail may be intentionally degraded. A simpler pulse shape, e.g. rectangular pulse, which takes only 4 points to calculate, may be used instead. This allows capturing the influence of finite pulse width – while ignoring the specific pulse shape.

### 3.4 Cross-verification

The solutions described in Sections 3.1 and 3.3 should give identical results if both are correct. These are plotted at six example sets of parameters in Figure 2. Comparison between the heating values $\theta$ in the numerical and analytical case shows nearly-identical results. Note that, for simplicity, the pulse was considered infinitesimal. The only minor deviations are caused by round-off errors and limited number of terms of series in the current implementation of the analytical algorithm. The numerical solution used a total $N = 30$ grid points.

### 4 Enhancing the problem statement

Both the diathermic and the coupled conductive-radiative problem statements are lacking in two important aspects:

- In the diathermic model, the Biot number $\text{Bi}(r)$ in the term $\eta\text{Bi}(r)(\theta_y=0 - \theta_y=1)$ is the same as in the heat loss term $\text{Bi}(r)\theta_y=0$. This only allows heat loss via infrared radiation. However, significant convective cooling has recently been reported [16] in a number of LFA measurements. Likewise, the value of emissivity $\varepsilon$ entering the expression for radiative fluxes $q(y)$ in the coupled conductive-radiative problem is also calculated based on the Biot number;

- The heat source term $\Phi(F_o)$ is always assumed to be localised at the front surface of the sample. This assumption breaks down if: the integrity of the graphite coating is breached e.g. by scratching, a conductive path between the front and rear faces exists, or light is scattered and reflected [20] onto
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Figure 2 Comparison of analytical and numerical solutions of (3a) – (3d) for six example sets of parameters: (1) – Bi = 0.01, \( \eta = 0.2 \); (2) – Bi = 0.9, \( \eta = 0.4 \); (3) – Bi = 0.9, \( \eta = 0.8 \); (4) – Bi = 5.0, \( \eta = 0.0 \); (5) – Bi = 5.0, \( \eta = 0.1 \); (6) – Bi = 5.0, \( \eta = 1.0 \). In the numerical case, a rectangular pulse width equal to 1/10,000 of the characteristic time \( l^2/a \) is assumed.

the rear sample surface. In fact, parasitic reflections have recently been discovered in both benchtop and free-standing LFA setups with different optical arrangements.

To address these two aspects, two alternative problem statements are proposed.

4.1 Revised diathermic model

A modified diathermic model is proposed as follows:

\[
\frac{\partial \theta}{\partial F_0} = \frac{\partial^2 \theta}{\partial y^2}, \quad 0 < y < 1, \quad F_0 > 0, \quad (19a)
\]

\[
\left. \frac{\partial \theta}{\partial y} \right|_{y=0} = Bi \cdot \theta_{y=0} + \eta Bi_{(r)} (\theta_{y=0} - \theta_{y=1}) - \Phi(F_0) \zeta, \quad (19b)
\]

\[
\left. \frac{\partial \theta}{\partial y} \right|_{y=1} = -Bi \cdot \theta_{y=1} + \eta Bi_{(r)} (\theta_{y=0} - \theta_{y=1}) + \Phi(F_0) [1 - \zeta], \quad (19c)
\]

\[
\theta(F_0 = 0) = 0, \quad (19d)
\]

where \( 0 < \zeta \leq 1 \) is the fraction of the pulse energy absorbed at the sample front surface. At \( \zeta < 1 \), a \( \sim [1 - \zeta] \) fraction of the pulse is deposited at the rear surface. Note this is somewhat similar to an expression previously proposed by Hahn et al. [8]. There, a source term combined two Dirac delta functions at
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$y = 0$ and $y = 1$. In contrast, (19b) and (19c) are formulated for an arbitrary pulse shape $\Phi(Fo)$. Therefore, the model takes full advantage of the pulse diode data recorded by the instrument. Additionally, a convective Biot number is introduced, $Bi_c = h L / \lambda$, where $h$ is a convective coefficient determined by the gas medium where the sample is immersed. This adds up to the radiative heat losses in the first term of (19b) and (19c) so that $Bi = Bi_c + Bi_r$.

4.2 Revised coupled conductive-radiative model

The coupled system of equations may also be modified to account for convective losses and stray light. These modifications are straightforward and done by complete analogy to the above. The first term in (1b) and (1c) should be replaced by $(Bi_r + Bi_c) \cdot \theta_{y=0}$ and $-(Bi_r + Bi_c) \cdot \theta_{y=1}$ respectively. Additionally, the heat source [last term in (1b)] should read as $-\Phi(Fo) \zeta$. An additional $\Phi(Fo) [1 - \zeta]$ term should be added in (1c).

5 Example applications

In the examples below, PULsE v1.95 [21, 22] was used to process the experimental data.

5.1 Polycrystalline alumina (PCA)

A Netzsch LFA 467 HT with a xenon pulse source ($Q \approx 0.5$ J) was used to measure a $l = 2.5$ mm, $\phi 12.6$ mm alumina sample. The sample front and rear faces were coated with a Kontakt Chemie Graphit 33 spray. Data on thermal properties $C_p$ and $\rho$ were preloaded in the processing software. This helped imposing realistic constraints on $Bi_r$. Prior to the parameter search, the initial values of $Bi_r$ at each temperature were calculated assuming $\varepsilon = 0.8$, which corresponds to pyrolitic graphite.

The diathermic model (19a) – (19d) was used to process the data. A significant contribution of the convective heat losses $Bi_c$ is observed in Figure 3 (a). This served to compensate insufficient radiative losses at low temperatures limited by an emissivity value $\varepsilon < 1$. Note that, at $T < 1200$ K, the initial curve segment due to sample transparency was less pronounced, preventing accurate estimation of the parameters $\varepsilon$ and $\eta$. The diathermic coefficient, $\eta$, displayed in Figure 3 (b) was below 1/2. As explained in Section 2.2, this might be interpreted as the influence of scattering. Finally, the thermal diffusivity variation with temperature was observed to be in reasonable agreement with a previous experiment by Hofmeister [23], see Figure 4. That latter experiment used a 1 mm alumina sample with a reflective coating applied on its surface and an implementation of Mehling’s model to process the data.

A more detailed coupled model was also employed to analyse the data collected on the PCA sample. The model used reference values for the refractive index in the infrared region ($n \approx 1.5-1.7$ [24]) of PCA. The variation of the output parameters with temperature is displayed in Figure 5. The emissivity,
Figure 3 Parameters extracted from the LFA measurements of a PCA sample using the coupled model with separate convective and radiative heat losses: (a) heat loss parameters $B_i(r)/l$ and $B_i(c)/l$; (b) diathermic coefficient, $\eta$. Error bars displayed for a 95% confidence level.

Figure 4 Temperature evolution of thermal diffusivity calculated with the revised diathermic and coupled models in PCA and synthetic sapphire. The reference diffusivity data is plotted according to [23] for PCA $(\parallel c)$. 
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Figure 5 Parameters extracted from the LFA measurements of a PCA sample using the model (19a) – (19d): (a) heat loss parameters Bi(r) and Bi(c); (b) Planck number NP; (c) Optical thickness τ0; (d) Scattering albedo ω0. Error bars displayed for a 95% confidence level.

ε, [Figure 5 (d)] and the convective heat losses, Bi(c), [Figure 5 (a)] were in agreement with the values estimated independently with the diathermic model. At the same time, the coupled model could also be used to infer the Planck number NP [Figure 5 (b)], optical thickness τ0 [Figure 5 (c)] and scattering albedo ω0 [Figure 5 (d)]. A Cornette-Schanks phase function was used to describe scattering [25] with an initial assumption on the free parameter g = 0 corresponding to Rayleigh scattering [26]. The results indicate a fairly constant optical thickness averaging at τ0 = 0.65, corresponding to an absorptivity value of 2.6 cm−1. Thermal diffusivity values are a few percent lower compared to the results of the diathermic model, see Figure 4. The deviation increases at higher temperatures where the contribution of radiative transfer quantified by the NP number becomes higher.

Although these parameter estimates seemed reasonable, the radiative contribution was weakly pronounced in thick samples, possibly contributing to a larger error margin. An example is shown in Figure 6 where radiative transfer
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Figure 6  Example LFA curve for a PCA sample at $T_0 = 1173$ K processed using a diathermic model with and without convective losses. A constraint $\varepsilon < 1$ has been imposed, requiring convective losses to accurately describe the heating curve. Additionally, the result for the coupled model is also plotted, showing it is required to reproduce the initial curve segment.

is only seen to affect the initial curve segment. Finally, both models are compared in terms of how well they fit the data. An example time-temperature profile at $T_0 = 1173$ K is shown in Figure 6. It is noted that a model with only radiative heat losses (dashed curve) does not describe the data correctly. While the diathermic model is able to capture the qualitative shape of the curve, it shows a deviation at the beginning of the curve. In this case, a coupled model is shown to produce a closer match to the data.

5.2 Synthetic sapphire

Raw LFA data on a $l = 1.181$ mm synthetic sapphire sample previously reported in [7] was analysed with the revised models. The calculation details are the same as in Section 5.1. This specific dataset was produced in an experiment where imperfect sample coating could be partially responsible for a sharp laser-induced peak on the time-temperature profile. Therefore, this dataset was useful to demonstrate the feasibility of a model with variable $\zeta$, see
Figure 7 Example LFA curve for a synthetic sapphire sample at $T_0 = 1682$ K processed using a coupled model considering only front face heating ($\zeta = 1$) and an additional contribution of the rear face ($\zeta < 1$). The initial temperature drop, partially attributed to the coating quality, is best reproduced at $\zeta \approx 0.97$.

Figure 7. Calculations with the coupled model resulted in parameter values shown in Figure 5. The closest match to the detector curves was achieved at a scattering albedo of unity, $\omega_0 = 1$, and a $\zeta \approx 0.97$. On the other hand, the diathermic model showed an even better match to the data, producing a sharper peak after the pulse [Figure 7]. However, as discussed in Section 2.2, the applicability of the diathermic model in case of strong absorption combined with scattering is questionable.

Analysis of diffusivity results in Figure 4 indicates an apparent discrepancy between the diathermic and coupled models in the sapphire sample at $T_0 > 1500$ K. At first, it looks surprising that a good agreement between the simplified and complex models is observed at lower temperatures – especially since scattering was estimated to be very strong in a broad temperature range [Figure 5 (d)]. However, the value of the Planck number at $T_0 < 1500$ K, estimated at $N_P > 3.5$ [Figure 5 (c)], causes a lower magnitude of the error introduced into calculations with a simplified model. As the temperature decreases, the relative contribution of the radiative fluxes in the computed $\theta(Fo)$ profile becomes lower, and significant errors in determining the radiative
fluxes lead to minor consequences in terms of the calculated temperature profile. Therefore, even a crude radiative transfer model might yield acceptable accuracy of measuring the diffusivity, $a$, at high $N_P$ values.

5.3 Opaque Zr sample with $\zeta < 1$

A Netzsch LFA 467 HT with a xenon pulse source was used to measure a $l = 0.2$ mm Zr-1% Nb sample. The sample pre-heated to 100 °C on a hot plate was then fully coated (including side surfaces) with a Kontakt Chemie Graphit 33 spray. This resulted in a sample fully enveloped in the graphite ‘thin shell’, promoting circumferential heat fluxes. A rapid pathway for heat transfer bypassing the sample material opens up. When the difference in diffusivities of the sample material and the graphite coating is large, temperature variations due to the coating occur at the time scale of the pulse. Effectively, this results in a rear-surface heat source. This is just one example of rear-face heating. In most LFA setups using a xenon light source, the pulse delivery system creates a diverging light beam. Stray light [20] is produced as the beam interacts with internal parts of the sample chamber. This light diverges from the standard optical path and may be absorbed at the rear face — under favourable conditions. Such conditions include IR transparency of high-temperature insulating materials e.g. the alumina protective tubes in a microheater setup. The latter may be opaque at a wavelength of $10 − 12$ μm characteristic to thermal radiation at room temperature. At the same time, it displays a high transmittance for waves shorter than $4$ μm, corresponding to temperatures above ca. 700 °C. As the transmittance increases, a larger flux of stray light might reach the sample rear face. To account for these complications, a modified set of boundary conditions include the $[1 − \zeta]\Phi(Fo)$ term.

In samples characterised by a larger $l^2/a$ value (cf. Figure 6), the flash seems instantaneous. In that case, it is difficult to tell the influence of $\zeta < 1$ from the scattering peak intrinsic to the material under study. Note $\zeta(T_0)$ is not a material property. Instead, it is a function defined by the geometrical and optical properties of the instrument and the sample coating. To validate a model with $\zeta < 1$, data on both the sample heating and the pulse power profiles are required. Example LFA measurements with $[1 − \zeta]$ ranging from 0.001 to 0.01 are shown in Figure 8. The shape of the ‘hump’ correlates with the pulse diode signal shown in Figure 8 (b), as may be expected from (19c). The results indicate that even a small fraction of the initial pulse power lost in form of stray light may result in a significant deviation from the classical behaviour of the LFA curve. The same may be expected in coated transparent media.

6 Discussions

The applicability of the diathermic model [3, 8] has been assessed. With the trivial exception of an opaque material ($\eta = 0$), it is shown to be strictly valid only for non-scattering, optically thin materials, with $\eta$ given by (4).
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Figure 8 Example dataset for a Zr-1%Nb sample, showing a ‘hump’ in the detector signal (a) and the corresponding pulse power profile (b). Markers correspond to raw data points and model solutions are plotted in solid lines. The hump may be accurately modelled with the $\Phi(Fo)[1 - \zeta]$ term, where $\zeta$ changes from 0.9988 to 0.9845.

Under certain assumptions, it may be considered approximately valid in case when scattering is present, resulting in $\eta$ given by (8). A constraint $\eta > 1/2$ must be imposed, if no scattering is present, according to (4). Logically, in the $0 < \eta < 1/2$ region, the model is approximately valid at best.

To calculate the pulse-corrected time-temperature profile $\theta(Fo)$, either (15) with (12a) – (12b) and (13a) – (13c) or the numerical solution described in Section 3.3 may be used. Concerning the analytical case, it is argued that using the transcendental equations (12a) – (12b), similar to those proposed by Hahn et al. [8], should be preferred. A different form of those equations considered by Mehling et al. [3] may be numerically less stable. As the transcendental equations are solved numerically, a potential source of error lies is the root-finding routine. Choosing a correct routine is of prime importance, as errors in finding the roots may jeopardize chances of success in calculating the $\theta(Fo)$ curve, see Section 3.2.1. In this regard, the implementation based on the ZEROIN algorithm by Brent and Dekker is recommended (see the Supplementary Material). On the other hand, the numerical solution described in Section 3.3 may be advantageous due to a more straightforward pulse correction, see Section 3.3.2. Additionally, the finite-difference scheme has the distinct advantage of relying on purely algebraic relations (18a) – (18c). Its accuracy is controlled by only two parameters, $h$ and $\tau_f$. Either the analytical or the numerical approaches, when implemented correctly, result in identical time-temperature profiles, see Figure 2.

In a revised treatment for transparent samples in LFA [Section 4], the convective $Bi_{(c)}$ and radiative $Bi_{(r)}$ heat losses are separated. Interestingly, this is somewhat similar to the problem statement proposed in [10]. When the specific heat and density data is available, this allows calculating $Bi_{(r)}$ prior
to the optimisation runs either using a standard emissivity of the graphite coating or imposing a $\varepsilon < 1$ constraint. Additionally, a breach of the coating causing flash-throughs or reflections due to stray light may be modelled by including an additional $\zeta$ parameter. Then, the rear-face ‘parasitic’ heating is proportional to $(1 - \zeta)$. Including this factor helps processing datasets with a sharp peak originating from the stray light or from a conductive circumferential path connecting the front and rear faces. An example treatment is shown in Figure 7.

Example measurements with LFA instruments in samples where absorption combined with scattering is significant showed a progressive error as the parameter $N_P$ decreased e.g. Figure 4. Apparently, this is caused by the simplifications of the diathermic model. Normally, the error introduced in the diffusivity, $a$, values is below 3% at $N_P \geq 3.5$. Since this error is of the same order as the expected accuracy of LFA measurements, the diathermic model may be used for data processing at these $N_P$ values. The relative weight of the radiative transfer is inversely proportional to $N_P$, requiring the use of a more detailed (coupled [7]) model to avoid larger errors in determining the diffusivity values, $a$. The latter were shown to be in the excess of 10% in PCA and sapphire samples at high temperatures. Note that, in case of the coupled model, the radiative fluxes entering (1a), (1b) and (1c) depend on the emissivity value. The variation of emissivity with other parameters is expressed as $\varepsilon = \text{Bi}(r)aC_p\rho/(4\sigma_0T_0^3l)$. In the example measurements, especially for the vertically-arranged LFA setup, a $\text{Bi}(r)$ corresponding to $\varepsilon = 1$ is unable to fit the cooling segment of the LFA datasets. To maintain sensible parameter values, it becomes necessary to introduce the convective losses, $\text{Bi}(c)$. This allows fitting the cooling segment of the curve without altering the $\text{Bi}(r)$ value. Additionally, introducing $\zeta$ may be required to avoid over-estimating the role of scattering in the coupled model.

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