Computer Assisted Methods in Engineering and Science, **22**: 279–287, 2015.

# Numerical data processing from a laser flash experiment on thin graphite layer

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In this paper, the methodology for determination of the out-of-plane thermal diffusivity (TD) of a thin graphite layer deposited onto a substrate of known properties is presented. The developed methodology resulted in combined experimental-numerical procedure enabling investigation of the properties of thin layer deposits. The procedure involves the experimental data acquisition during the laser flash tests, and next the numerical processing of the collected data using the heat conduction problem solution and the nonlinear least square parameter identification approach. Two last steps produce a certain inverse heat conduction problem that is formulated and numerically solved for a three-layer specimen. The procedure has been successfully tested while processing the real experimental data from investigation of flake graphite layers. This proved the effectiveness of the methodology in providing quantitative data on the TD of thin layers of relatively poor conductors deposited onto a highly conductive substrate.

**Keywords:** inverse analysis, finite element method (FEM), multilayer heat transfer, thin layer diffusivity, laser flash data processing.

## 1. INTRODUCTION

Flake graphite is deposited onto the specimen surface in a laser flash [21] investigations of the thermal diffusivity (TD) in order to improve the absorbance of laser flash energy and to enhance the infrared thermal response at contactless measurements [19]. However, the heat capacity and thermal resistance of the graphite coating can affect the result of measurements [8]. Several studies have been performed in order to evaluate the measurement error resulting from the application of the surface treatment. The influence of graphite coatings and the specimen thickness on the results of flash TD measurements of selected materials was determined analytically and experimentally in [1, 7, 13]. The adequate error correction procedures were proposed in several studies [5, 8, 12]. Nevertheless, there is still a lack of reliable data on thermal properties of thin graphite layers. This is mostly because standard TD measurement techniques fail to investigate thin layers. Thermophysical properties of thin layers can differ from properties of bulk material and thus the investigation of such structures usually requires a non-standard approach. Up to date, several attempts have been made to accommodate the standard TD measurement methods for thin layer studies, mostly in view of thermal barrier coatings [2, 7]. He et al. [10] estimated the TD of thermal barrier coating by fitting the analytical model response to the experimental data. Maglic [16] developed a two-dimensional laser flash method for measuring the TD of deposed solid thin films. McMasters [15] presented a method for investigation of thin film TD from laser flash experimental data. The proposed method involved inverse problem solution. The numerical simulation of the heat transfer in three-layer specimen was applied to solve the direct problem. Chen [5] proposed a data reduction method in which the thermal response of a sample is fitted with calculated curve for the numerical simulation by nonlinear least-square regression. The method was applied for determination of the TD and the thermal resistance of a coating deposited on a substrate of known thermal properties [6]. Terpiłowski [25] applied a modified laser flash method to investigation of plastics in a three-layer structure. None of these studies resulted in reliable TD data of the flake graphite. Such data are necessary for a more precise determination of the TD measurement uncertainty when applying the laser flash apparatus [1, 12, 17].

In the course of presented studies, a procedure has been developed for determination of the out-of-plane TD of the flake graphite layer. The procedure combines laser flash measurements and numerical modelling with the inverse problem solution. The flash experiments are performed on a three-layer sample that is a carrier/base specimen made of a highly conductive material, whose upper and bottom side are covered with graphite. The associated direct heat conduction problem in the course of inverse procedure is formulated and solved utilising the finite element method (FEM) COMSOL software. This procedure was successfully tested already in the numerical simulation presented in [22, 23]. The current paper covers the issue of the procedure testing on real experimental data and the estimation procedure of uncertainty analysis.

## 2. Method

While developing the methodology, the following criteria were set forth: i) the signal data from standard thermal diffusivity measurements will be utilised, ii) the investigated material layers will be applied onto a specimen made of material of known properties, iii) the TD will be determined from the thermal response of a three-layered specimen (graphite layer – carrier specimen – graphite layer), iv) the complementing experimental parameters will be determined within the same procedure, and v) it will be possible to apply the developed method for investigation of various coatings. In order to match these criteria a dedicated multi-parametric estimation procedure was developed. Due to the complex heat transfer in a three-layer specimen that is exposed to a surface flash excitation it is not so convenient to apply typically used analytical models [11, 14]. Therefore, an inverse problem solution including fitting the experimental laser flash data with the results of numerical simulation was proposed. The direct problem was formulated for a three-layer specimen and solved using the commercial FEM COMSOL software. Multi-parametric estimation was performed using the Levenberg-Marquardt algorithm implemented into the MATLAB software programme.

### 2.1. Inverse problem formulation

In the proposed methodology, the TD of a thin layer is estimated by fitting the numerical model response $T = (T)_{i=1,\dots,I}$ to the appropriate experimental data series $Y$. The solution of the analysed inverse heat problem is based on minimisation of the ordinary least-squares norm in the form:

$$S(P) = \sum_i [Y_i - T_i(P)]^2, \tag{1}$$

where $S$ – is a sum of square error, $P$ – is a vector of unknown parameters, $T_i(P)$ – is the estimated temperature at time $\tau_i$, $Y_i$ – is temperature measured at time $\tau_i$. The unknown experimental parameters were: $a$ – thermal diffusivity of the graphite layer, $h$ – heat transfer coefficient and $q$ – heat flux. Therefore, a vector of three parameters was estimated:

$$P = [a, h, q]. \tag{2}$$

The solution of direct problem in the form of vector of temperatures $T_i$ is calculated applying the FEM COMSOL software. The multi-parametric estimation procedure was performed using the Levenberg-Marquardt algorithm [18]. A detailed description of sensitivity coefficient analysis and numerical testing procedure of the developed algorithm can be found in [23].

### 2.2. Direct problem

The direct problem solution included a simulation of heat transfer in a three-layer specimen excited with a pulse heating on one of its surfaces (see Fig. 1). The typical specimen in laser flash test has a disk shape. Therefore, in order to reduce computational cost, a two-dimensional (2D) axisymmetric numerical model has been applied. The developed model accounts for convective heat losses and for the thermal contact resistance at the interface between the base specimen and graphite. The convective heat transfer at the specimen surfaces was provided with the use of *heat flux* assisted by the COMSOL software boundary condition, while the contact resistance was obtained with a *thin, thermally conductive layer* on internal boundary. A more detailed description of the model can be found in [23].

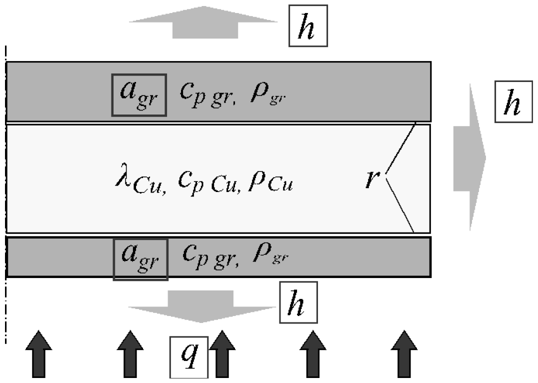

**Fig. 1.** A scheme of a three-layer model developed for direct problem solution. The estimated parameters are circled with squares.

### 2.3. Experiments

The experiments were performed with the use of a Netzsch Laser Flash Apparatus (LFA) 457. During every single measurement the bottom surface of the specimen is heated with the laser flash and the thermal response signal is collected from the opposite upper side by the infrared (IR) detector. In the course of standard measurements, the TD (or the apparent/effective TD; see Fig. 2) is determined from the acquired data by using a dedicated Netzsch Proteus software.

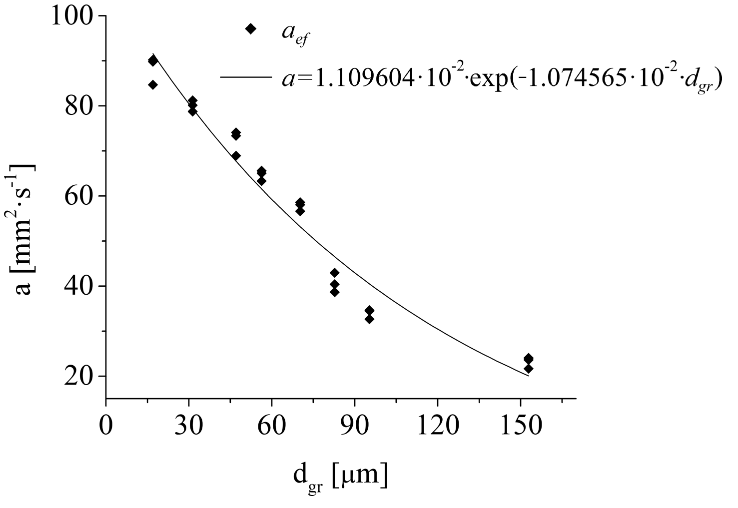

**Fig. 2.** The graphite layer thickness effect on the effective thermal diffusivity – single flash results from the Netzsch Proteus software (measurements performed at 30°C).

The commercial software provides a variety of models for correction of error resulting from the finite pulse time [4, 24], convective and radiative heat losses from the specimen surface [9], and non-uniformities in the flash special distribution and non-uniform initial temperature distribution in the specimen. Usually, the experimental data is processed accounting for many different models, and finally the so-called best fit result of the TD is proposed. In our study, this final best fit result was most frequently obtained from Cowan's model (see [9]); therefore, it was decided that this type of results would be taken arbitrarily as the representative results of the apparent TD measurement. These results are shown in Fig. 2. However, the apparent TD of the composite three-layer specimen could not be recalculated for a thin-layer material TD so easily. Therefore, in the presented work the signal from detector is processed with a designedly developed numerical procedure. The investigated graphite layer was deposited on specimens made of copper in order to maximise the influence of coating on a sample's thermal response. Four similar carrier specimens were used: each of them of a disk shape and approximately 1 mm thick and of 12.5 mm diameter. The graphite layer was deposited on copper carrier specimens by aerosol spraying. The side exposed to laser flash was covered with a layer of equal thickness of approximately 7 μm, whereas the side facing the detector was covered with layers of different thickness for every specimen. The various thicknesses of graphite layers ranged from 7 to 150 μm. The measurements were performed at temperatures 30, 50, 100, 175, 225 and 300°C. At every single reference temperature, the flashes were repeated three times. The raw measurement data for temperature 30°C are shown in Fig. 3.

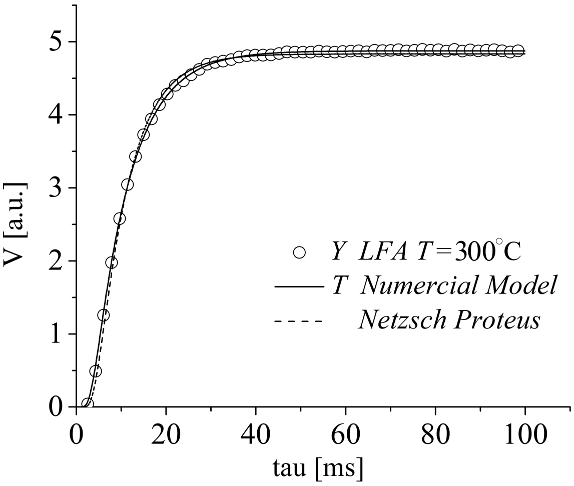

**Fig. 3.** Typical result of fitting the numerical simulation result to the experimental LFA signal for high temperature measurements; additionally, a fitting curve from the Netzsch Proteus software is shown.

## 3. RESULTS OF FLAKE GRAPHITE THERMAL DIFFUSIVITY ESTIMATE

Determination of the thermal diffusivity of the graphite layers was performed at three stages: a) acquiring the thermal response of three-layer specimen to a laser flash excitation with LFA, b) determination of the effective TD of investigated specimen with the Netzsch Proteus software, and c) numerical processing of the collected data with the developed procedure. The data processing procedure is in accordance with the following scheme: i) raw experimental signal from IR detector is used as an input of the inverse procedure, ii) the heat conduction problem is solved for a given set of parameters $P$ with the FEM COMSOL software, iii) the simulated thermal response $T(P)$ is fitted to the experimental data $Y$, iv) if the following stopping condition for the numerically defined [22] parameter $\varepsilon$ is not satisfied:

$$\sum_i [Y_i - T_i(P)]^2 < \varepsilon \tag{3}$$

a new value of vector $P$ is set using a Levenberg-Marquardt nonlinear least square algorithm, and v) if the condition (3) is satisfied than the procedure is stopped, the actual values of the vector $P$ are considered as the best estimates and the TD $a$ is determined. An exemplary result of the data fitting corresponding to final iteration of the procedure is presented in Fig. 3. The multi-parametric estimation algorithm was developed in MATLAB and a coupling between MATLAB and COMSOL was used for iterative change of the FEM parameters and the simulation. Prior to main calculations, the performance of the developed algorithm and that of the commercial software were compared. The effective values of TD of three-layered specimen was determined with the developed procedure and the Netzsch Proteus software. The comparison of the obtained results (see Table 1) proved good performance of the presented numerical data processing procedure.

**Table 1.** Comparison of effective thermal diffusivity determined for a three-layered sample. The best fit result according to the Netzsch Proteus software is marked with asterisks*.

| Netzsch Proteus 6 | | | Developed numerical data processing procedure |
|---|---|---|---|
| Adiabatic model* | Parker model | Cowan model | |
| $a_{ef}$ [mm$^2$·s$^{-1}$] | | | |
| 36.967 | 35.927 | 33.737 | 37.226 |

### 3.1. Typical results of identification procedure

The developed procedure was tested in the course of numerical simulations [23]. During these tests a full study of the model sensitivity was performed, the effect of the thermal response noise on the estimation result was investigated, and the adequate value of the stopping parameter was determined. The simulation results proved correctness and effectiveness of the developed procedure. Thus, the same methodology as described in [23] has been applied to process the real experimental data. While conducting the present studies, the TD of graphite coating of various thicknesses has been determined. The LFA measurements were performed with the temperature ranging from 30 to 300°C. The final results of the infrared detector output signal processing are presented in Table 2 and in Fig. 4. The TD measurements were supplemented by investigations of the graphite coating density and heat capacity. Similar investigations were performed for the copper specimen. All studies resulted in determination of the TD of the graphite layer and thermal conductivity, but a discussion of these results is out of the scope of this paper. The appropriate details are provided in [22]. The TD of the out-of-plane flake graphite decreases with increasing temperature, independently of the layer thickness. Experimental investigations and numerical calculations have been supplemented by a detailed error analysis. The analysis, as was expected, resulted in a relatively high uncertainty values (see the following section). Nevertheless, it does not affect reliability of the obtained TD data.

**Table 2.** The thermal conductivity and thermal diffusivity of a 59 µm thick graphite layer determined by developed procedure.

| T [°C] | $\lambda$ [W·m$^{-1}$·K$^{-1}$] | $a$ [mm$^2$·s$^{-1}$] |
|---|---|---|
| 30 | 1.049 | 1.51 |
| 50 | 1.396 | 1.85 |
| 100 | 0.793 | 0.88 |
| 175 | 1.203 | 1.07 |
| 225 | 1.174 | 0.86 |
| 300 | 1.150 | 0.77 |

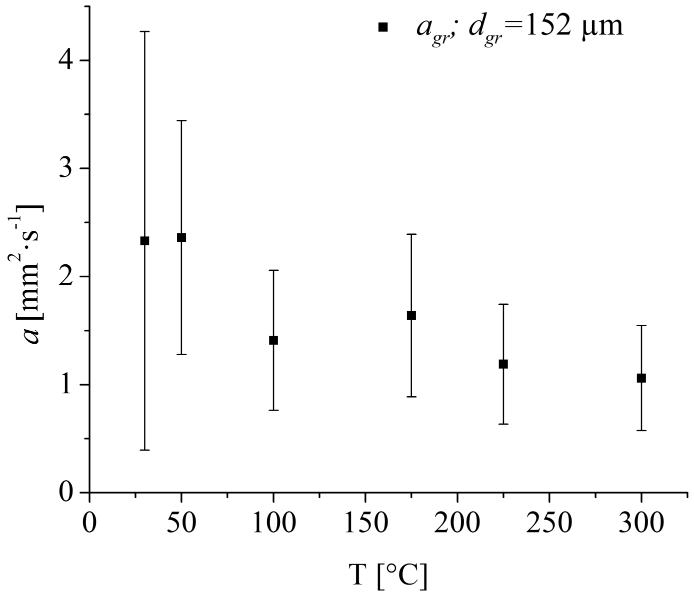

**Fig. 4.** Typical results of the experimental data processing in the form of the flake graphite thermal diffusivity (the studied coating was 152 µm thick).

### 3.2. Uncertainty analysis

There are many error sources influencing the final result because of the complexity of the measurement and parameter estimation. Nevertheless, at least three types of error sources can be distinguished and analysed individually. The first type are error sources typical for laser flash method and they include errors caused by imperfect matching of experimental conditions by the model (see, e.g., [4, 11, 17, 20, 24]). The second type are error sources strictly connected to the proposed three-layer structure geometry. This particular concept increases the needs for the procedure input data. Except the knowledge of the base specimen geometrical parameters, the additional knowledge on some thermophysical properties of the investigated materials is needed too. The third type are errors that arise from inverse processing of the acquired signals. The results of a detailed measurement uncertainty study has been presented and discussed in [22]. Among others, the uncertainty analysis accounted for: i) finite time of pulse heating, ii) mismatch between the model rectangular distribution and the real changes in time of a laser flash intensity, iii) non-uniformities of the surface heating by a laser flash, iv) uncertainty in determination of copper specimen thickness, v) uncertainty in determination of graphite layer average thickness, vi) graphite layer material anisotropy, vii) thermal expansion, viii) non-uniform distribution of graphite coating, ix) the uncertainty of specific heat in the thermal conductivity calculations, and x) estimation procedure errors. The sources of measurement errors from i) to iii) have been discussed in [4, 9, 24]. According to the previous analyses, they would not much affect the final result in the discussed case of the thin layer TD investigations. Nevertheless, during the numerical procedure testing this was additionally confirmed. The errors in determination of the layer thickness (iv), the layer material graphite density (v) and the heat capacity (ix) contribute much to the total uncertainty [20, 22]. However, they are not intrinsic error sources of the developed inverse procedure itself. As far as the anisotropy (vi) is concerned, the final effect on the uncertainty revealed to be minor – mostly because of dominating one-dimensional (1D), axial heat conduction within the investigated specimen. Crucial effects were observed when analysing estimation procedure errors. This is because of an increasing IR noise while the measurement reference temperature (i.e., the initial specimen temperature) is decreasing. Within the frame of the inverse procedure optimisation and the uncertainty analysis, a detailed sensitivity study was also performed. The analyses concluded that the most significant effects are: the error in the graphite layer thickness evaluation and the cumulative error in the inverse processing of the investigated thermal response (see, e.g., Fig. 3). Those factors result in more than a 10% uncertainty of the final result in the out-of-plane effective TD of the flake graphite coating layer.

Since nonintrusive measurement of the thickness of the graphite coating deposited onto a carrier specimen was not possible, a gravimetric measurement was performed. The thickness was calculated from the mass of the coating and the density of graphite. Therefore, the errors in determination of mass have contributed to the coating thickness determination. As a result, a total error of layer thickness determination was estimated to be 18%.

In the analysis of the estimation procedure errors, the confidence intervals for all the estimated parameters were determined with a 99% confidence level:

$$P - 2.58\sigma_p \le \widehat{P} \le P + 2.58\sigma_p, \tag{4}$$

where $\sigma_P$ is a vector of standard deviations for estimated parameters [18]. Confidence intervals turned out to be dependent on the temperature. This is because of the IR detector sensitivity characteristics – low temperature signals exhibited much more scatter than high temperature ones. Typical results of the confidence interval evaluation are shown in Table 3.

**Table 3.** Confidence intervals for estimated parameters at high and low temperatures of thermal diffusivity investigation.

| T = 30°C | T = 225°C |
|---|---|
| $P = [1.4838\quad 4.8140\quad 5.2880\cdot 10^6]$ | $P = [0.6823\quad 6.0924\quad 4.9925\cdot 10^7]$ |
| $0 < a < 6.1843$ | $0.6736 < a < 0.6909$ |
| $0 < h < 9.9898$ | $3.0553 < h < 9.1295$ |
| $3.8767\cdot 10^6 < q < 6.6993\cdot 10^6$ | $4.8948\cdot 10^7 < q < 5.0902\cdot 10^7$ |

The estimation procedure uncertainty depends not only on the temperature but also on the graphite coating thickness. This problem is discussed in details in [22]. Nevertheless, this contributes to the total uncertainty of about 60% at low temperatures and decreases with increasing temperature to less than 1%, but only in the case of thick coatings studies (see [22]).

Because of a complex interdependence of different error source effects it is not possible to determine one representative value characterising the TD determination uncertainty. Every single experiment should be treated individually. Nevertheless, the total uncertainty contributes much to final results as shown in Fig. 4. One of the possible methods of improving the accuracy is to increase the number of repeated single measurements. By performing an additional analysis focused on the layer thickness effect on the thermal diffusivity estimation sensitivity, it was proved that the sensitivity decreases with a decreasing TD of the substrate specimen. For this reason, the substrates with a high TD need to be used.

## 4. SUMMARY

In this paper, a numerical data processing that resulted in determination of the out-of-plane TD of thin graphite coating has been discussed. The processed data come from laser flash experiments performed on a three-layer specimen. Because of limitations of the experimental methodology and limitations of the standard software, direct TD calculations are not possible. For this reason, a dedicated procedure combining the laser flash experiments, numerical modelling of the heat conduction in the investigated specimen and inverse processing of the experimental vs. numerical data has been developed. In a course of performed numerical and experimental tests, the developed procedure has proved to be effective in providing reliable data while investigating thin layers of relatively poor conductors deposited onto a highly conductive substrate. The out-of-plane TD of graphite coating at room temperature was estimated to be equal to $1.6 \pm 0.9\ \mathrm{mm^2{\cdot}s^{-1}}$. The flake graphite TD decreases with an increasing temperature.

In contrast to the inverse problem solutions utilising the analytical models [10, 16] and regularisation techniques [3], the application of the FEM modelling allows to freely modify the geometry of the direct problem solution. For this reason, the developed methodology can facilitate investigations of a variety of layered structures, including thermal barrier coatings.

## ACKNOWLEDGMENTS

The experimental instrumentation of this research was completed within the frame of the 558/FNiTP/691/2010 project.

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