Nonlinear Effects in the Laser Flash Thermal Diffusivity Measurements

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Abstract

Numerical solution of nonlinear heat conduction equation is used to analyze nonlinear effects in the laser flash experiment, when the thermophysical parameters of the sample depend on the temperature. Parameter estimation technique is proposed to determinate the temperature dependence of the thermal diffusivity from a response curve. Computer generated data as well as a real experimental data were used to demonstrate the technique.

Key words: Numerical algorithm, nonlinear heat conduction, temperature distribution

1 Introduction

In the laser flash method [1] one surface (at $x = 0$) of a small disc shaped sample of thickness L is irradiated by a laser pulse and resulting temperature rise at opposite surface ($x = L$) is used to calculate the thermal diffusivity α of the sample material.

Existing data reduction methods for calculation of thermal diffusivity from the temperature rise of the sample are based on assumption that the thermophysical parameters - heat capacity c and thermal conductivity λ (and also thermal diffusivity $\alpha \equiv \lambda/c$) are constants independent of temperature T within the temperature range of the a flash experiment. One (or two) dimensional linear heat conduction equation

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2}, \tag{1}$$

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with initial and boundary conditions relevant to the experiment is solved and the thermal diffusivity is calculated by fitting the experimental temperature rise to the appropriate analytical solution of the Equation (1). For most of materials, temperature range and a final temperature rise $\lesssim 1$ K, the assumption of constant thermophysical properties is valid and the results of the thermal diffusivity determination are usually within a couple percent of claimed experimental uncertainty of the flash method.

Use of short and powerful laser pulses to measure very thin samples lead to a temperature rise much greater than than a couple degrees K assumed for a perturbation type experiment. The assumption that the temperature rise of the sample is not very high is not longer valid. If the heat capacity $c(T)$ and the thermal conductivity $\lambda(T)$ varies with temperature then temperature distribution in sample is found by solving the nonlinear heat conduction equation:

$$c(T)\frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(\lambda(T)\frac{\partial T}{\partial x}\right). \tag{2}$$

Equation (2) will be solved numerically in this paper for a constant heat capacity $c(T) = c_0$ and thermal conductivity as a function of temperature

$$\lambda(T) = \frac{a_0}{a_1 T + 1}, \tag{3}$$

where $a_0, a_1 > 0$ are positive constant parameters. The effect of temperature dependent $\lambda(T)$ on flash method diffusivity measurement was analyzed in [2] where it was found that nonlinearity can be neglected up to certain degree given by the value of the parameter a_1. We will show that these parameters can be determined from the response curve in the laser flash experiment using a parameter estimation technique. Computer generated, as well as real experimental data will be used to demonstrate the usage of the proposed procedure.

2 Numerical Solution

Equation (2) has been solved numerically using an implicit difference scheme [3]. Sample thickness L is divided into $N = 21$ elements. The sample is initially in equilibrium state at temperature T_0. Heat pulse is assumed to be instantaneous (at $t = 0$) and its energy is absorbed in the first element, rising its temperature to T_1. Sample boundaries are adiabatically insulated.

Temperature $T_{i,m+1}$ of the i-th element ($i = 1, 2, 3, \ldots, N$) at the time $t_{m+1} = (m + 1)\Delta t$, $m = 1, 2, \ldots$ is given by a system of equations:

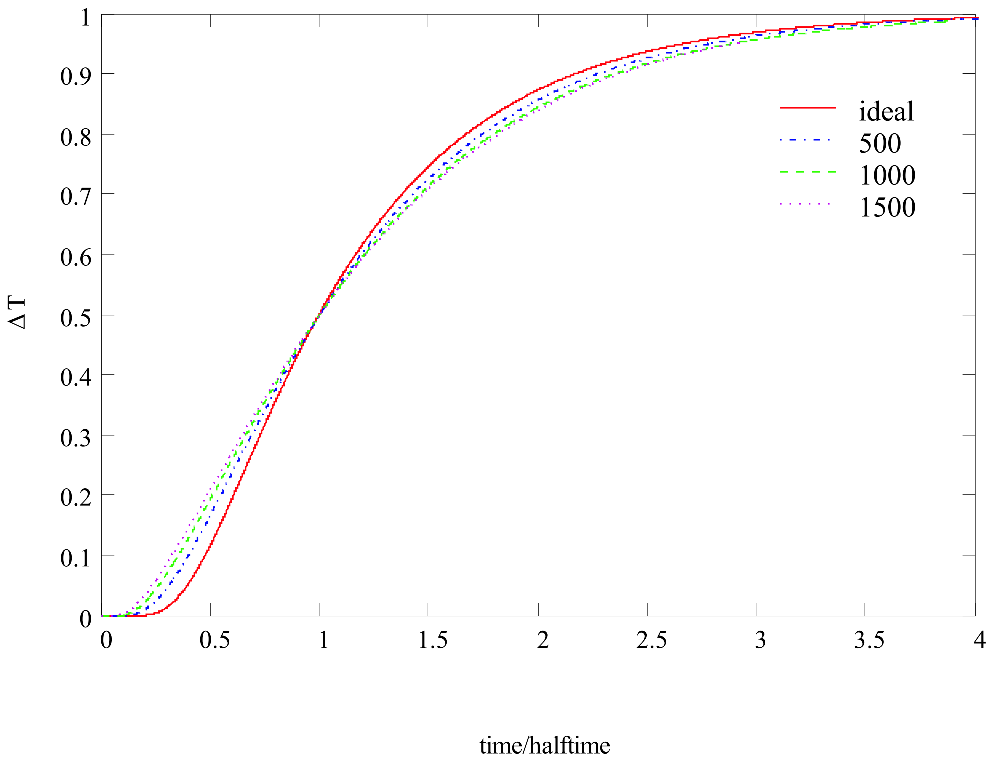

Fig. 1.
Nonlinear temperature rise for different initial temperature T_1 of the sample front surface.

$$\begin{aligned} T_{1,m+1} &= T_{1,m} + 2Fo_r(T_{2,m+1} - T_{1,m+1}) \\ T_{i,m+1} &= T_{i,m} + Fo_l(T_{i-1,m+1} - T_{i,m+1}) + Fo_r(T_{i+1,m+1} - T_{i,m+1}) \\ &\text{for } i = 2, 3, \ldots, N-1 \\ T_{N,m+1} &= T_{N,m} + 2Fo_l(T_{N-1,m+1} - T_{1N,m+1}) \end{aligned} \quad (4)$$

where

$$\begin{aligned} Fo_l = \frac{\Delta t \lambda_l}{c \Delta x^2}, \qquad Fo_r = \frac{\Delta t \lambda_r}{c \Delta x^2} \\ \lambda_l = \frac{2\lambda_{i-1}\lambda_i}{\lambda_{i-1} + \lambda_i}, \qquad \lambda_r = \frac{2\lambda_{i+1}\lambda_i}{\lambda_{i+1} + \lambda_i} \end{aligned} \quad (5)$$

and λ_i is is the thermal conductivity of i-th element. A standard iterative algorithm was used to solve Equations (4).

Nonlinear temperature rise $V(L,t)$ at $x = L$ was calculated for a temperature dependence of $c(T) = c_0$, where c_0 is constant value of heat capacity at T_0,

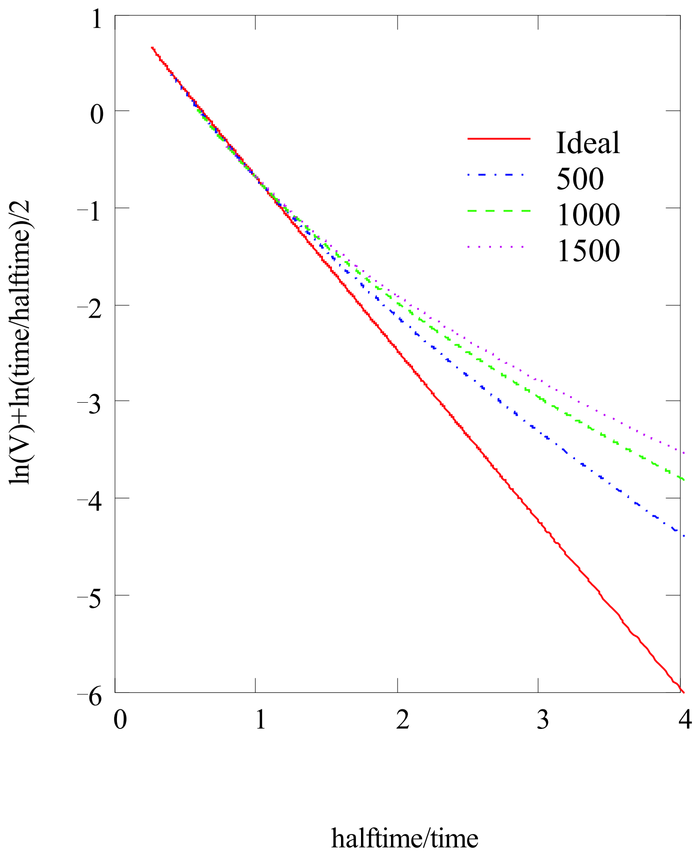

Fig. 2.
Function $(\ln(V) + \ln(t/t_{1/2})/2)$ versus $t_{1/2}/t$ for a different T_1.

and $\lambda(T)$ given by Equation (3). Since the heat capacity is constant, the temperature dependence $\alpha(T)$ will be similar to $\lambda(T)$.

Figure 1 shows the nonlinear temperature rise $V(L,t)$ as a function of time for various initial temperatures T_1 of the first element. The curves are normalized to a new steady temperature after the pulse and time is normalized to halftime value $t_{1/2}$. (Halftime is a time needed for the temperature at $x = L$ to rise to half of its new steady state value after the pulse.) Ideal curve for constant values of $c(T) = c_0$ and $\lambda(T) = \lambda_0$ is also presented in Figure 1. Shape of the nonlinear curves differs from the ideal curve. Generally, the nonlinear curves lead the ideal one in the first half of their rise and lag behind the ideal one in the second half. Curves for higher T_1 rise more slowly, than those for lower T_1. The shape distortion is more noticeable for the curves with higher T_1.

The differences between the ideal and nonlinear curves are more visible in Figure 2, where a plot of $(\ln(V) + \ln(t/t_{1/2})/2)$ versus $t_{1/2}/t$ is presented for a different initial temperature T_1. Ideal curve, given by

$$V_i(L,t) = \frac{L}{\sqrt{\pi \alpha t}} \sum_{n=0}^{\infty} \exp\left[-\frac{(2n+1)^2 L^2}{4\alpha t}\right], \tag{6}$$

is a straight line with a slope $-L^2/4\alpha t_{1/2} \doteq 1.80$. The deviations from the ideal curve shape increase with T_1.

The differences in shape between the ideal and nonlinear curve make it impossible to match nonlinear curve with an ideal one using a constant value of the thermal diffusivity. In principle [4] no single effective temperature T_e can be found for $c(T_e)$ and $\lambda(T_e)$ to describe the solution of nonlinear equation. Experimental nonlinear curves can be normalized and apparent thermal diffusivity value can be calculated from the halftime $t_{1/2}$ using Parker's formula

$$\alpha = 0.139 \frac{L^2}{t_{1/2}}, \tag{7}$$

but the results will be a function of T_1 (laser energy), as it was observed on graphite samples in [5].

3 Parameter Estimation Technique

Determination of temperature dependent thermophysical properties from the measured temperature responses is a *coefficient inverse problem* and many numerical and analytical methods were proposed to solve this problem (see e. g. [6]. In this paper, we describe a new simple parameter estimation technique to determinate unknown coefficients of the temperature dependent thermal conductivity (diffusivity) given by Equation (3) from a measured temperature response in the flash method.

Sensitivity study of nonlinear response curve showed that its sensitivity coefficients [7] (partial derivatives with respect to a_0, a_1, T_0 and T_1 respectively) are linearly independent, so the coefficients $\vec{\beta} \equiv (a_1, a_2, T_0, T_1)$ can be found simultaneously.

Ordinary least square procedure (Fortran package ODRPACK [8]) was used to find the unknown parameters $\vec{\beta}$ from

$$\min_{\vec{\beta}} \sum_{i=1}^{n} [f_i(t_i; \vec{\beta}) - y_i]^2, \tag{8}$$

where $f_i(t_i; \vec{\beta})$ is the temperature point at time t_i calculated using the numerical solution given by Equations (4) and (t_i, y_i), $i = 1, 2, 3, \ldots, n$ are the points of the temperature response curve (observed data).

4 Results and Discussion

Five different sets of temperature rise were generated by a computer in order to demonstrate proposed parameter estimation technique. In this example the stability and accuracy of the technique are tested. Set 1 was generated using: $L = 0.002$ m, $c_0 = 10^6$ J/(m^{3}K), $a_0 = 100$ W/(mK), $a_1 = 0.05$ K^{-1}, $\Delta t = 10^{-6}$ s, $T_0 = 0$ °C and $T_1 = 500$ °C. The sets 2, 3, 4 and 5 were generated using Set 1 data with different level of noise added to the temperature points. Superimposed noise imitates experimental errors and was generated using random number generator. The sets differ from each other by noise to signal ratio.

The results of parameter estimations are listed in Table 1. The reproducibility and accuracy of the calculated parameters is relatively high, even for Set 5 with the highest noise to signal ratio. The differences between estimated and exact value of the parameter are $< 1\%$ in all cases. Standard deviation (SD) values of a_0 are $< 1\%$ and SD values of a_1 are $< 2.5\%$ of the estimated value.

Real experimental data has to be carefully examined before the proposed parameter estimation technique is applied. Similar distortion can be caused by e. g. finite pulse time effect when the pulse duration is comparable with the halftime value or by a nonlinearity of the temperature detector used in the experiment. Repeated measurements using different laser energy, different pulse duration, or using different sample thicknesses have to be conducted to identify the presence of nonlinearity in response curves.

A strong dependence of the apparent thermal diffusivity on laser pulse energy for POCO ZXF-5Q graphite sample at room temperature was reported in [5]. The apparent values of the diffusivity were lower for higher laser pulse energy. A plausible explanation was found in a fact that the thermal diffusivity of graphite decreases with temperature and the dependence is stronger at room temperature than at elevated temperatures.

Our laser flash experiments with a graphite foam samples at temperatures around room temperature also showed temperature rise curves distortions similar to the nonlinear curves plotted in Figure 1. Typical temperature rise of graphite foam sample ($L = 2.01 \cdot 10^{-3}$ m, $c_0 = 6.86 \cdot 10^5$ J/(m^{3}K)) after the pulse was about 8 °C. Apparent thermal diffusivity was $\alpha = 1.19 \cdot 10^{-4}$ m^2/s. After finite pulse time correction [9], the thermal diffusivity value was $\alpha = 1.45 \cdot 10^{-4}$ m^2/s. The results of our parameter estimation technique were: $T_0 = 99.03$ °C, $T_1 = 427$ °C, $\alpha(T_0) = (2.16 \pm 0.15) \cdot 10^{-4}$ m^2/s and $a_1 = 0.093$ K^{-1}. The value of the thermal diffusivity at T_0 calculated using the parameter estimation technique seems to be more realistic than the corrected apparent value. On the other hand, the value of parameter a_1 indicates that the thermal diffusivity decreases with temperature more rapidly than was found in the

Table 1
Results of parameter estimation.

Set No.	Ratio N/S	T_0 °C	T_1 °C	a_0 W/(mK)	SD W/(mK)	a_1 1/K	SD 1/K
1	0	0.00	500.00	100.00	0.00	0.05000	0.00000
2	0.008	0.00	499.96	99.87	0.20	0.04981	0.00025
3	0.017	0.00	499.93	99.74	0.43	0.04962	0.00057
4	0.025	0.00	499.89	99.60	0.64	0.04943	0.00086
5	0.042	0.00	499.85	99.47	0.86	0.04925	0.00114

experiment. The response curve was distorted mainly due to the finite pulse time effect.

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