International Journal of Thermophysics, Vol. 25, No. 4, July 2004 (© 2004)

Nonlinear Effects in Laser Flash Thermal Diffusivity Measurements[1]

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The numerical solution of the nonlinear heat conduction equation is used to analyze nonlinear effects in the laser flash method, when the thermophysical parameters of the sample depend on the temperature. A parameter estimation technique is proposed to determine the temperature dependence of the thermal diffusivity from a response curve. Computer generated data, as well as real experimental data, were used to demonstrate the reliability of the technique.

KEY WORDS: laser flash method; nonlinear heat conduction; numerical algorithm; 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 the resulting temperature rise at the opposite surface ($x = L$) is used to calculate the thermal diffusivity α of the sample material.

Existing data reduction methods for calculation of the thermal diffusivity from the temperature rise of the sample are based on the 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 flash experiment. The one-(or two-) dimensional linear heat conduction equation

[1]Paper presented at the Fifteenth Symposium on Thermophysical Properties, June 22–27, 2003, Boulder, Colorado, U.S.A.
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$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} \tag{1}$$

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 Eq. (1). For most materials, the 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.

The use of short and powerful laser pulses to measure very thin samples leads to a temperature rise much greater than a few kelvins assumed for a perturbation-type experiment. The assumption that the temperature rise of the sample is not very large is no longer valid. If the heat capacity $c(T)$ and the thermal conductivity $\lambda(T)$ vary with temperature, then the temperature distribution in the 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 a thermal conductivity that depends on temperature as

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

where $a_0, a_1 > 0$ are positive constant parameters. The effect of the temperature dependent $\lambda(T)$ on flash method thermal diffusivity measurement was analyzed in Ref. 2 where it was found that nonlinearity can be neglected up to a level determined 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 data, as well as real experimental data, will be used to demonstrate the reliability of the proposed procedure.

2. NUMERICAL SOLUTION

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

The temperature $T_{i,m+1}$ of the ith 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:

$$\begin{aligned} T_{1,m+1} &= T_{1,m} + 2Fo_{\rm r}(T_{2,m+1} - T_{1,m+1}), \\ T_{i,m+1} &= T_{i,m} + Fo_{\rm l}(T_{i-1,m+1} - T_{i,m+1}) + Fo_{\rm r}(T_{i+1,m+1} - T_{i,m+1}), \\ &\qquad \text{for } i = 2, 3, \ldots, N-1, \\ T_{N,m+1} &= T_{N,m} + 2Fo_{\rm l}(T_{N-1,m+1} - T_{1N,m+1}), \end{aligned} \tag{4}$$

where

$$\begin{aligned} Fo_{\rm l} &= \frac{\Delta t \lambda_{\rm l}}{c\Delta x^2}, \qquad & Fo_{\rm r} &= \frac{\Delta t \lambda_{\rm r}}{c\Delta x^2}, \\ \lambda_{\rm l} &= \frac{2\lambda_{i-1}\lambda_i}{\lambda_{i-1} + \lambda_i}, \qquad & \lambda_{\rm r} &= \frac{2\lambda_{i+1}\lambda_i}{\lambda_{i+1} + \lambda_i} \end{aligned} \tag{5}$$

and λ_i is the thermal conductivity of the ith element. A standard iterative algorithm was used to solve Eq. (4).

The nonlinear temperature rise $V(L,t)$ at $x = L$ was calculated for a temperature dependence of $c(T) = c_0$, where c_0 is a constant value of heat capacity at T_0, and $\lambda(T)$ is given by Eq. (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-state temperature after the pulse and time is normalized to the halftime value $t_{1/2}$. (The halftime is the time needed for the temperature at $x = L$ to rise to half of its new steady-state value after the pulse.) The ideal curve for constant values of $c(T) = c_0$ and $\lambda(T) = \lambda_0$ is also presented in Fig. 1. The 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 slower 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 Fig. 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. The ideal curve, given by

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

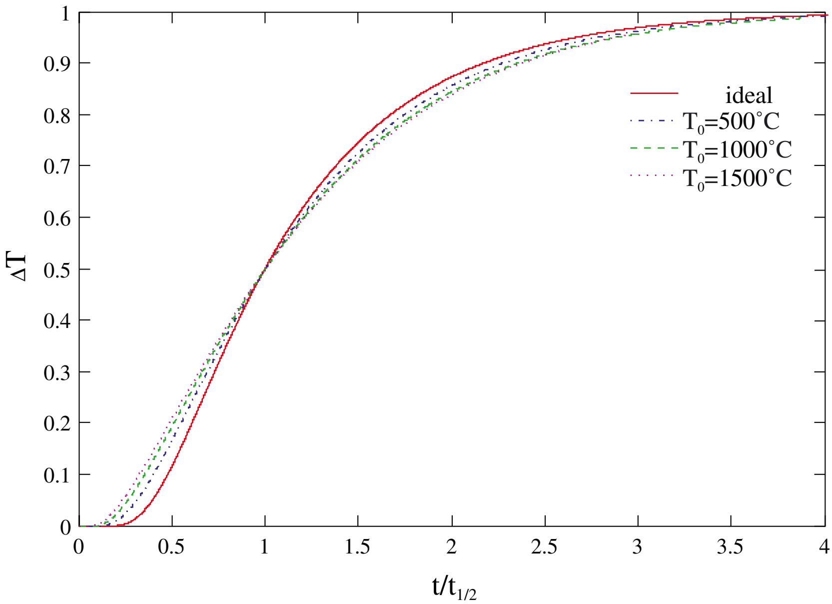

Fig. 1. Nonlinear temperature rise for different initial temperatures T_1 of the sample front surface.

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 curves make it impossible to match a nonlinear curve with an ideal one using a constant value of the thermal diffusivity. Theoretically [4], no single effective temperature T_e can be found for $c(T_e)$ and $\lambda(T_e)$ to describe the solution of the nonlinear equation. Experimental nonlinear curves can be normalized and apparent thermal diffusivity values 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 was observed on graphite samples [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

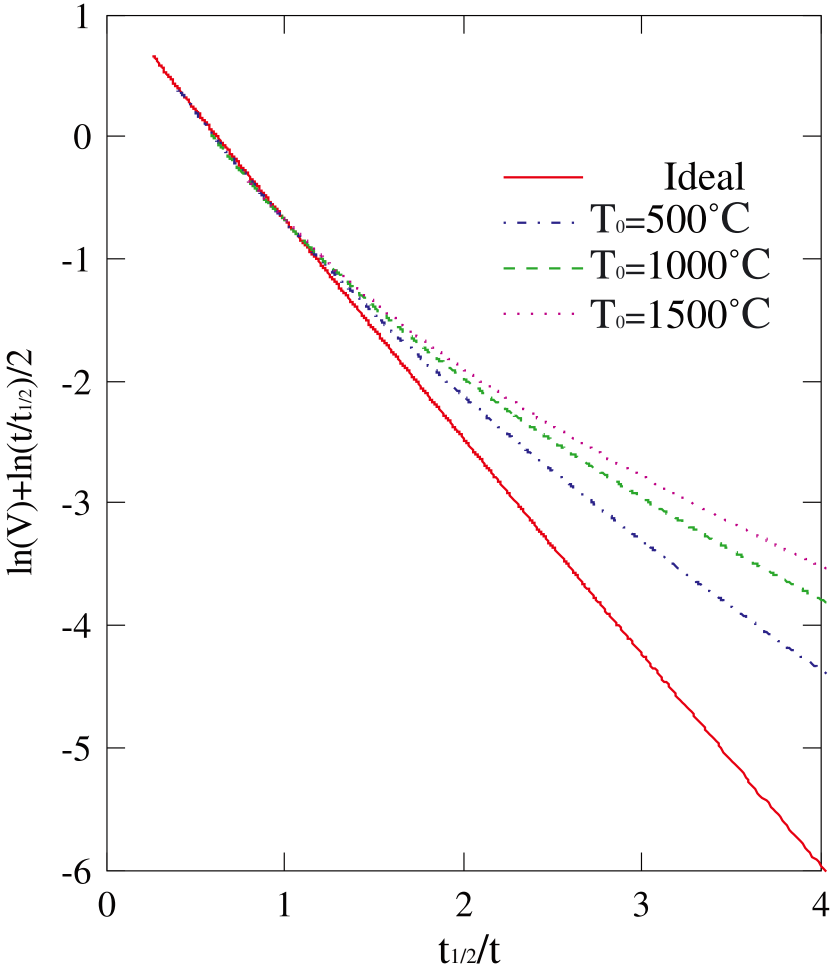

Fig. 2. Function $(\ln(V)+\ln(t/t_{1/2})/2)$ versus $t_{1/2}/t$ for different initial temperatures T_1.

problem (see, e.g., Ref. 6). In this paper, we describe a new simple parameter estimation technique to determine unknown coefficients of the temperature-dependent thermal conductivity (diffusivity) given by Eq. (3) from a measured temperature response in the flash method.

A sensitivity study of the nonlinear response curve showed (see Fig. 3) 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.

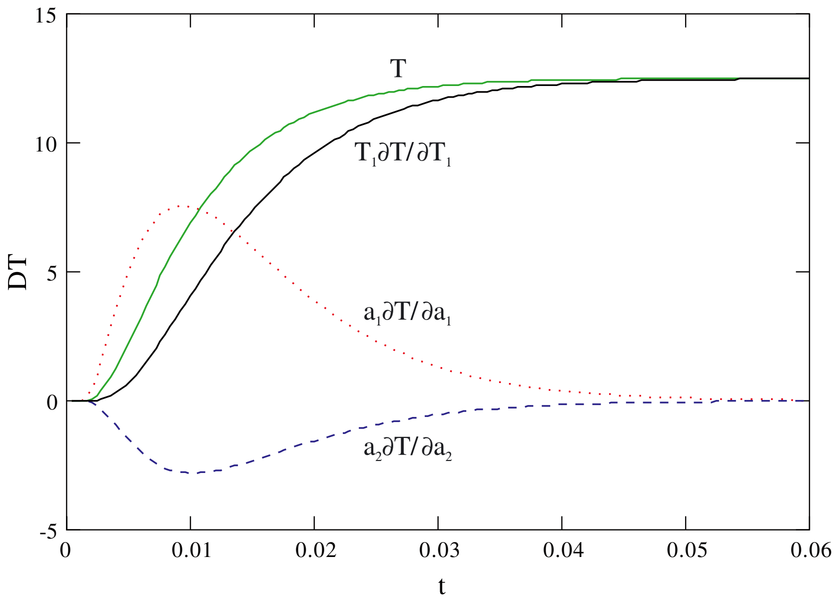

Fig. 3. Sensitivity coefficients as functions of time.

Ordinary least-squares procedures (Fortran package ODRPACK [8] were used to find the unknown parameters $\vec{\beta}$ from

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

where $f_i(t_i; \vec{\beta})$ is the temperature point at time t_i calculated using the numerical solution given by Eq. (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 the 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, $c_0 = 10^6\,\mathrm{J \cdot m^{-3} \cdot K^{-1}}$, $a_0 = 100\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$, $a_1 = 0.05\,\mathrm{K^{-1}}$, $\Delta t = 10^{-6}$ s, $T_0 = 0\,^\circ$C, and $T_1 = 500\,^\circ$C. The sets 2, 3, 4, and 5 were generated using Set 1 data with different levels of noise added to the temperature points. Superimposed noise imitates

Table I. Results of Parameter Estimation

Set No.	Ratio N/S	T_0 (°C)	T_1 (°C)	a_0 ($W \cdot m^{-1} \cdot K^{-1}$)	SD ($W \cdot m^{-1} \cdot K^{-1}$)	a_1 (K^{-1})	SD (K^{-1})
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

experimental errors and was generated using a random number generator. The sets differ from each other by the noise-to-signal ratio.

The results of parameter estimations are listed in Table I. The reproducibility and accuracy of the calculated parameters are relatively high, even for Set 5 with the highest noise-to-signal ratio. The differences between estimated and exact values 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 values.

Real experimental data must be carefully examined before the proposed parameter estimation technique is applied. Similar distortion can be caused by, e.g., a 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 energies, different pulse durations, or using different sample thicknesses have to be conducted to identify the presence of nonlinearity in the response curves.

A strong dependence of the apparent thermal diffusivity on laser pulse energy for a POCO ZXF-5Q graphite sample at room temperature was reported in Ref. 5. The apparent values of the diffusivity were lower for a higher laser pulse energy. A plausible explanation was 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 graphite foam samples at temperatures around room temperature also showed temperature rise curve distortions similar to the nonlinear curves plotted in Fig. 1. A typical temperature rise of a graphite foam sample ($L = 2.01 \times 10^{-3}$ m, $c_0 = 6.86 \times 10^5$ $J \cdot m^{-3} \cdot K^{-1}$) after the pulse was about 8°C. The apparent thermal diffusivity was $\alpha = 1.19 \times 10^{-4}\, m^2 \cdot s^{-1}$. After a finite pulse time correction [9], the thermal diffusivity value was $\alpha = 1.45 \times 10^{-4}\, m^2 \cdot s^{-1}$. The results of our parameter estimation technique were: $T_0 = 99.03$°C, $T_1 = 427$°C, $\alpha(T_0) = (2.16 \pm 0.15) \times 10^{-4}\, m^2 \cdot s^{-1}$, 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 experiment. The response curve was distorted mainly due to the finite pulse time effect.

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