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Analysis of thermal diffusivity by parameter estimation in converging thermal-wave technique

Sok Won Kim [a,*], Jong Chul Kim [b], Sang Hyun Lee [b]

[a] Department of Physics, University of Ulsan, Ulsan 680-749, Republic of Korea
[b] Division of Physical Metrology, Korea Research Institute of Standards and Science, Taejon 305-600, Republic of Korea

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Abstract

The in-plane thermal diffusivity of several kinds of metal foils was measured by the converging thermal-wave technique. This is a typical technique which can obtain the thermal diffusivity by taking the temperature evolution at the center of pulsed annular laser beam when the beam irradiates the surface of the samples. The thermal diffusivity is normally calculated using the maximum time t_m or half-maximum time $t_{1/2}$ when the temperature evolution is half and maximum, respectively, however, the rapid temperature increase and the nonlinearity of the infrared detector in the earlier part, convection heat loss from the sample surface, and some times the low signal to noise ratio can produce errors. In this study, a nonlinear least-square regression was performed to estimate the optimal values of several separate parameters by fitting the data to the theoretical equation. The thermal diffusivity of the samples obtained from the optimal values of the estimated parameters was compared with the values from t_m and $t_{1/2}$, and the reference values, and it shows that the thermal diffusivity obtained by this parameter estimation technique agrees quite well with the reference data within 3.4% at maximum.

Keywords: Convection heat loss; Converging thermal-wave technique; Infrared detector; In-plane thermal diffusivity; Metal foil; Parameter estimation technique

1. Introduction

Recently, various kinds of thin-film or thin-plate materials have been developed in many fields, for example, for use in advanced microelectronic devices and in space and atomic technologies [1]. Also, the understanding of heat transfer and measurement of thermal diffusivity in those thin materials have become increasingly important in technological applications. There are many thermal diffusivity measurement methods, however, the laser flash technique developed by Parker et al. [2] has been widely used due to the need for rapid and simple measurements. In this technique, the thermal diffusivity was deduced from only one experimental point such as the maximum temperature time or half maximum temperature time, hence the accuracy was some times not so good. In order to solve this problem, several researchers [3,4] introduced curve fitting methods by which the entire experimental temperature history curve is fitted with the theoretical curve and obtained successful results.

However, it is limited to the through-plane thermal diffusivity of thin opaque samples of known thickness [5]. The converging thermal-wave technique is a kind of flash technique which can measure the in-plane thermal diffusivity by the measurement of temperature evolution at the front and rear center of a pulsed annular laser beam, when the laser beam irradiating the surface of the sheet samples and the thermal wave converge in the center [6–9].

The thermal diffusivity is conventionally obtained by checking the maximum temperature time [6,8] or half maximum temperature time [7,9]; however, the reduction of errors caused by the rapid temperature rise, the relatively wide range of the maximum time and sometimes noisy

* Corresponding author. Tel.: +82 52 259 2388; fax: +82 52 259 1693.
E-mail address: sokkim@ulsan.ac.kr (S.W. Kim).

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Nomenclature

c_p	specific heat at constant pressure (J kg^{-1} K^{-1})
E	energy liberated per unit thickness (J m^{-2})
p_{1-3}	parameters for estimation
R	radius of annular laser beam
S	sum of $[\theta(t)_i - T(t)_i]^2$
t	time (s)
t_{m}	maximum time (s)
$t_{1/2}$	half-maximum time (s)
$T_{\max}$	theoretically maximum temperature (arb. units)
$T_{1/2}$	half-maximum temperature, $T_{\max}/2$
$T(t)_i$	theoretically calculated temperature (arb. units)
T_0	zero level of detector output (arb. units)
Greek symbols	
α	thermal diffusivity (m^2 s^{-1})
λ	thermal conductivity
ρ	density (kg m^{-3})
$\theta(t)_i$	experimentally obtained temperature (arb. units)
Γ_j	normalized sensitivity coefficient

temperature evolution curve complicates the measurement [10]. Therefore, in spite of the same experimental data, the obtained thermal diffusivities show scattered values, which depend on the data analysis method.

In this work, the thermal diffusivities of several metallic foils were obtained by parameter estimation which is a powerful technique [11] that uses all the available data points and provides statistical means to analyze the experiment under the assumption of infinitely thin and infinitely short laser pulse. The proposed method makes it possible to reduce the several error sources mentioned above.

2. Analysis of converging thermal-wave model

2.1. Temperature evolution

In the case of an infinitely thin and infinitely short annular laser pulse of energy E liberated per unit thickness of a thin sample, the theoretical time-dependent temperature evolution $T(t)$ obtained at the center of the annulus of radius R, under the assumption that the finite pulse time effect and convection heat loss from the material surface is neglected, and in-plane propagation of the thermal-wave, is [12]

$$T(t) = \frac{E}{c_p \rho \pi R^2} \frac{R^2}{4\alpha t} \exp\left(-\frac{R^2}{4\alpha t}\right), \tag{1}$$

where $\alpha = \lambda/(c_p\rho)$ is the thermal diffusivity of the material, λ is the thermal conductivity, c_p is the specific heat at constant pressure, and ρ is the mass density. Such an expression can be used to evaluate the thermal diffusivity α from the experimentally observed temperature evolution curve.

Fig. 1 shows the theoretical curve obtained from Eq. (1). Lu and Swann [6] differentiated Eq. (1) with respect to t and obtained the maximum temperature time t_{m} when the temperature evolution becomes maximum and expressed the thermal diffusivity as $\alpha = R^2/4t_{\mathrm{m}}$. Cielo et al. [7] defined the half-maximum time $t_{1/2}$ when the temperature evolution becomes half-maximum, and they expressed the thermal diffusivity as $\alpha = R^2/(10.7t_{1/2})$. However, the errors caused by the rapid temperature rise in the earlier evolution curve, the nonlinearity of the detector, the relatively wide range of maximum time, and sometimes the noises from the low signal to noise ratio makes it complicate.

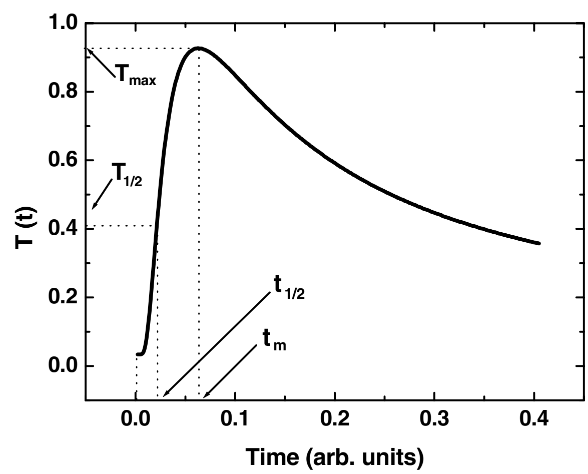

Fig. 1. Theoretical curve of temperature evolution and the definition of the characteristic points half-maximum time $t_{1/2}$ and maximum time t_{m}.

2.2. Parameter estimation (PE)

PE is a powerful technique that uses all the available data points and provides statistical means to analyze the experiment. The experimental data will be combined with the above model and used in a nonlinear sequential PE algorithm, NL2SOL developed by Dennis et al. [13,14], to determine the thermal diffusivity more effectively. The PE algorithm utilizes a method of least squares to minimize the sum, S, with respect to the desired parameters:

$$S = \sum_{i=1}^{N} [\theta(t)_i - T(t)_i]^2, \tag{2}$$

where $\theta(t)_i$ are the experimental data values and $T(t)_i$ are calculated values using the model given in Eq. (1). In order to apply PE to Eq. (1) we have to determine three unknown

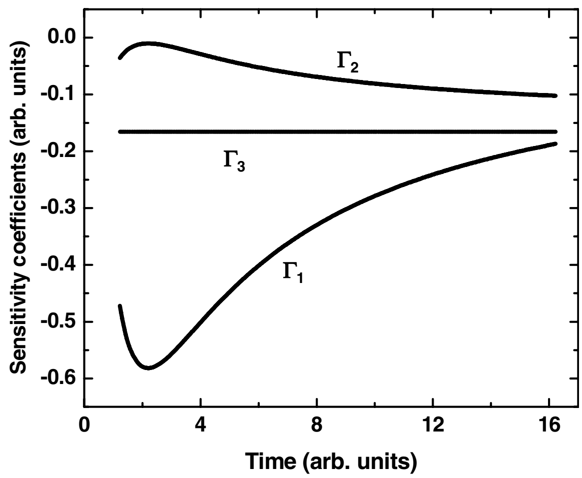

Fig. 2. Calculated sensitivity coefficients of the three parameters p_1, p_2 and p_3.

parameters p_1, p_2, and p_3, where $p_1 = \alpha$, $p_2 = E$, and $p_3 = T_0$ (zero level of detector output signal) as follows:

$$T(t)_i = \frac{p_2}{c_p \rho \pi R^2} \frac{R^2}{4p_1 t} \exp\left(-\frac{R^2}{4p_1 t}\right) + p_3. \quad (3)$$

The reliability of the estimated parameters obtained by using NL2SOL software depends on the sensitivity coefficient (SC) of each parameter [15]. The first derivative for the expression of temperature evolution with respect to the parameter is the SC, and when normalized to units of the amplitude, they appear as

$$\Gamma_j = p_j \frac{\partial T}{\partial p_j}, \quad (4)$$

where Γ_j is the normalized sensitivity coefficient, j is the index of parameters. The SCs for the parameters can provide a considerable amount of insight as to the adequacy of the model. If the model behaves similarly with a change in one parameter as it does with a change in another parameter, then the parameters are correlated, or dependent to some degree.

Fig. 2 represents the calculated SCs, Γ_j, for each parameter. The Γ_3 for p_3 is constant, but the Γ_1 and Γ_2 for p_1 and p_2, respectively show the quasi-symmetry relation and the Γ_1 decreases as Γ_2 increases and vice versa, however their amplitude is totally different. Therefore, we can say that p_1 and p_2 are slightly dependent to some degree, but p_3 is nearly independent on the others, and our model is relatively adequate.

3. Experiment

Fig. 3 is a schematic diagram of the experimental apparatus for the converging thermal-wave technique similar to the Lu and Swann [6] transmission method. The pulsed laser beam (duration: 0.8 ms, energy: 1.8 J) from an Nd:glass laser becomes annular when it passes through the combination of a converging lens and an axicon and

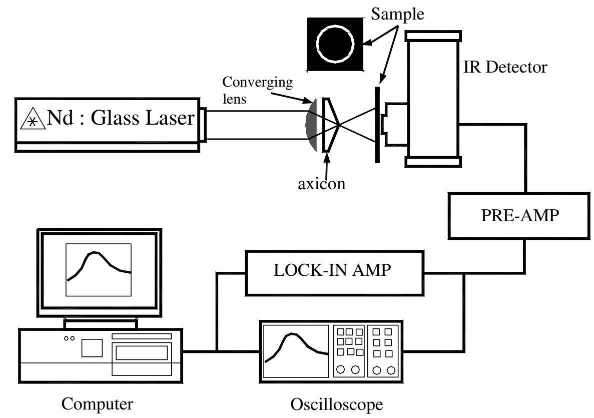

Fig. 3. Schematic diagram of experimental apparatus for converging thermal-wave technique.

Table 1
Sample descriptions

Specimen	Purity (%)	Thickness (μm)
Aluminum	99.5	50, 100
Copper	99.9	50, 100
Nickel	99.7	50, 100
Stainless steel 304	Cr: 18–20, Ni: 8–11, Mn < 2, Si < 1, balance Fe	50, 100

is focused on the sample surface. The reflectivity of the metal surface is higher than 80%, thus, in order to reduce that reflectivity the surface was coated with liquid graphite thin film whose thickness is less than 0.1 μm. The axicon was made of fused quartz, and its apex angle was 170°. The radius and width of the annular laser beam can be calculated by Snell's law using the values of the focal length of the converging lens, and the length from the lens to axicon. The converging time of the thermal-wave depends on the radius of the annular laser beam, so it must be maintained between 4 and 7 mm for optimum results [6].

The thermal-wave converted from the annular laser beam propagates in all directions, and the temperature evolution of the center is detected in the rear surface of the sample by an infrared HgCdTe detector. The temperature evolution was recorded by digital oscilloscope through a GPIB interface, and the thermal diffusivities were obtained by the PE.

The four kinds of metal foils measured were aluminum, copper, nickel, and stainless steel 304 whose thickness were 50 μm and 100 μm. Table 1 summarizes the sample materials. Also, to remove the effect of the diverging beam reflected from the edge of the sample, the sample was cut to 45 mm × 45 mm.

4. Results and discussion

Fig. 4 is the temperature evolution curves at the rear-side center of the annular laser beam after irradiating the laser beam on the aluminum sample whose thickness are 50 μm and 100 μm and the theoretical curves obtained by

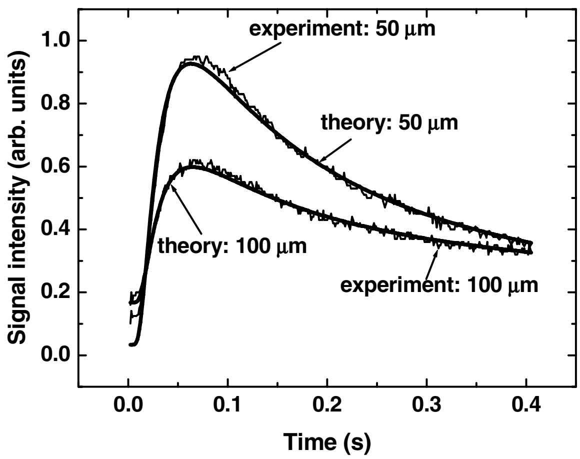

Fig. 4. Theoretically predicted curves by PE and the temperature evolution curves at the center of the annular laser beam on aluminum foil samples whose thickness are 50 μm and 100 μm.

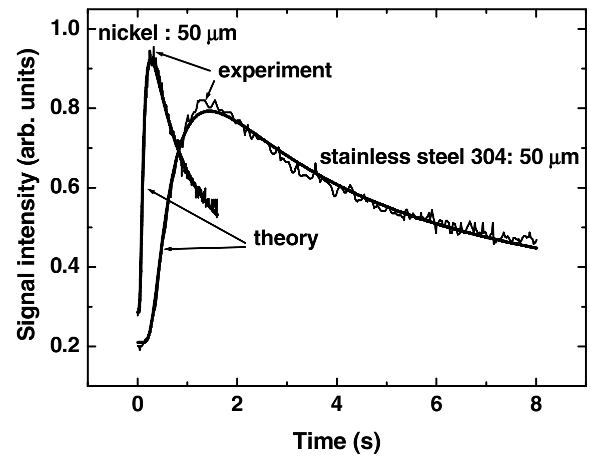

Fig. 6. Theoretically predicted curves by PE and the temperature evolution curves at the center of the annular laser beam on nickel and SS304 foil samples whose thickness is 50 μm.

the PE technique minimized the S value in Eq. (2). It shows that, immediately after the laser irradiation, the temperature increases rapidly and after the peak, it decreases slowly. It also shows that there are some noises even though the high energy pulse laser beam was used as a thermal source, therefore it is not easy to obtain accurately the thermal diffusivity by using the previous only one time point; that is a maximum time t_m or a half-maximum temperature time $t_{1/2}$.

The difference between the two curves is caused only by the different thickness (=different heat capacity) of the same material and the area under the 50 μm curve is approximately twice as that of 100 μm sample. The experimental and theoretical curves for 50 μm aluminum and copper samples, and for nickel and SS304 samples are shown in Figs. 5 and 6, respectively, however, those for the 100 μm samples were omitted because the all the trends are similar to the case of aluminum sample. The estimated values of parameters from all the experimental curves using Eq. (3) are shown in Table 2. The p_1 itself is a thermal diffusivity, p_2 is pulse energy per unit thickness, and p_3 is a base line voltage of the HgCeTe detector.

All the values of p_2 for 50 μm samples are not exactly but approximately twice as that of 100 μm samples. This is thought to be caused by the slightly different reflectance of the coated sample surfaces. Because of the drift of the base line voltage of HgCdTe detector caused by the high gain of the pre-amp, the value of p_3 varies from 0.034 to 0.288, however, the drift it is very slow, therefore it does not affect on the estimation values of other parameters of p_1 and p_2.

The obtained thermal diffusivities of the samples are shown in Table 3 with the values calculated by the equations of Lu and Swann [6], Cielo et al. [7], and reference data [16]. Thermal diffusivities obtained by the PE technique agree well with the reference data within 3.4% at maximum. Also, it shows that there is no dependence on thickness, therefore the assumption of one-dimensional propagation of thermal wave is effective in such metal foils whose thickness is up to about 100 μm. The pulse duration of the laser beam is 0.8 ms, and this is very short compared to several hundred milliseconds for the temperature evolution. Therefore, the finite pulse time effect can be neglected.

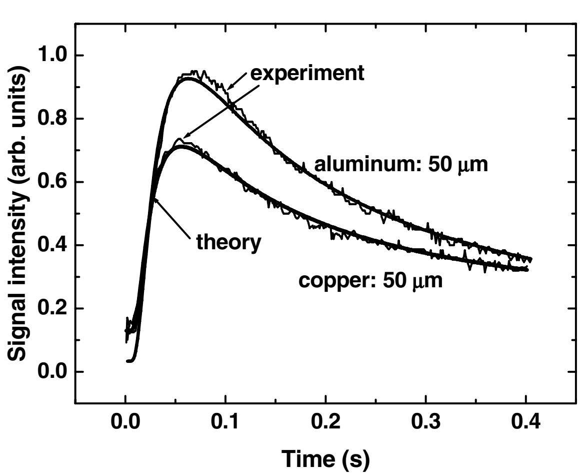

Fig. 5. Theoretically predicted curves by PE and the temperature evolution curves at the center of the annular laser beam on aluminum and copper foil samples whose thickness is 50 μm.

Table 2
The estimated values of the three parameters from the parameter estimation

Sample		Parameters		
Material	Thickness (μm)	p_1	p_2	p_3
Al	50	0.978	458.80	0.034
	100	0.964	221.22	0.167
Cu	50	1.130	437.01	0.130
	100	1.182	220.09	0.225
Ni	50	0.233	436.59	0.286
	100	0.231	226.88	0.288
SS304	50	0.0404	459.86	0.210
	100	0.0405	231.96	0.204

Table 3
Thermal diffusivities of several metal foil samples obtained by parameter estimation technique compared to other values from Lu's and Cielo's equations and with reference values

Sample		Thermal diffusivity (10^{-4} m^2 s^{-1})				Deviation between PE and reference (%)
Material	Thickness (μm)	PE	$\frac{R^2}{4t_m}$ [4]	$\frac{R^2}{10.7t_{1/2}}$ [7]	Reference value [16]	
Al	50	0.978	0.924	0.812	0.968	1.0
	100	0.964	1.000	0.812	0.968	−0.4
Cu	50	1.130	1.193	1.026	1.17	−3.4
	100	1.182	1.020	1.145	1.17	−1.0
Ni	50	0.233	0.242	0.206	0.229	1.7
	100	0.231	0.217	0.211	0.229	0.9
SS304	50	0.040	0.0471	0.0356	0.04	0.0
	100	0.041	0.0464	0.0375	0.04	2.5

The values obtained by the Lu and Swann equation deviate 16.7% at maximum from the reference values because the relatively wide range of the maximum time and noisy temperature evolution curve complicate the determination of t_m. For the case of SS304 sample, the deviation is larger than other samples because the slowly propagating thermal-wave causes the larger convection heat loss from the sample surface.

The diffusivity calculated by Cielo et al.'s equation shows about 16.1% lower values. These are caused by the fact that the nonlinearity of the infrared detector [17]; the $t_{1/2}$ obtained in the earlier part of the temperature evolution curve becomes longer than real time and it contains errors caused from the rapid evolution. In this experiment, the pulse energy is 1.8 J, and the amount of temperature increase at the center by this energy is about 12 K, therefore about 5% of nonlinearity of the infrared detector is contained in the data. These were verified by the fitting of experimental results to the theory.

The nonlinearity of the infrared detector which is more dominant in the earlier part of the temperature evolution curve decreases the thermal diffusivity and the convection heat loss produced in later part increases the thermal diffusivity, therefore in the PE technique which uses all the available data points, both effects act complementarily to each other and can reduce the errors.

At present, the problems of convection heat loss can be automatically cancelled out, therefore putting the samples into a vacuum chamber is not necessary. However, if the thermal diffusivity at higher temperature range will be needed, the sample must be put into a vacuum chamber in order to suppress the increasing convection heat loss.

5. Conclusion

The in-plane thermal diffusivities of several four kinds of metal foils whose thickness are 50 μm and 100 μm, obtained by the parameter estimation technique agrees well with the reference values within 3.4% at maximum, however the thickness dependence is not shown apparently. By comparison with the values obtained by the common technique which uses t_m or $t_{1/2}$, it can be concluded that the errors from the rapid temperature increase and the nonlinearity of the infrared detector in the earlier part, convection heat loss from the sample surface, and some times the low signal to noise ratio can be reduced remarkably.

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