Research Article

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Effect of radiative heat loss on thermal diffusivity evaluated using normalized logarithmic method in laser flash technique

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Abstract: The laser flash technique is a standard method to measure the thermal diffusivity of solid samples especially at high temperatures. To understand the reliability of thermal diffusivity evaluation at high temperature for solid samples with low-thermal-diffusivity values, we analyzed the effect of radiative heat loss using the logarithmic method. The results revealed that when the Biot number was 0.1, the deviation from the input thermal diffusivity value was approximately -1.6% . In addition, when an aluminum silicate (AS) sample was heated to 1,273 K, the maximum deviation was approximately -0.35% . In contrast, the difference between the input value and the thermal diffusivity evaluated by the halftime method when AS was heated to 1,273 K was approximately 2.38%. Thus, since the effect of radiative heat loss was found to be negligible, it is concluded that the normalized logarithmic method should be very useful for the thermal diffusivity analysis of low-thermal-diffusivity solid samples at high temperature.

Keywords: thermal diffusivity, solid sample, high temperature, radiative heat loss, laser flash technique, normalized logarithmic method

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1 Introduction

The laser flash technique is a standard method for thermal diffusivity measurements in solid materials, which are especially important at high temperatures [1,2]. In this technique, thermal diffusivity is determined by analyzing the temperature response of the rear surface of the sample just after flashing a laser pulse on the front of the sample. The easiest approximation for this analysis based on theoretical principles can be achieved through the so-called halftime method [3]. This analysis assumes ideal conditions based on the following criteria: (a) instantaneous irradiation with infinite pulse width, (b) uniform energy density for the laser beam, and (c) no heat loss from the sample during measurement. However, for real measurements, corrections are needed because of unavoidable nonideal conditions.

The thermal diffusivity evaluated by the halftime method is affected by radiative heat loss from the specimen surface, to a certain extent, at temperatures above 1,000 K. Some studies using the curve fitting method have been published [4,5]. Although this technique is the most conventional method for thermal diffusivity analysis [6], it is not suitable for the thermal diffusivity analysis of low-thermal-diffusivity materials such as oxide ceramics; this is because the time required for analysis when using the curve-fitting method is more than several seconds [7]. In the curve-fitting method, the entire experimental data set is fitted to a theoretical curve on the basis of Josell's analysis, which models the effect of radiative heat loss exactly [5]. The thermal diffusivity and Biot number are simultaneously determined by the curve-fitting method. In order to evaluate the uncertainty of the thermal diffusivity associated with this data analysis, the regions analyzed must include both the rising and the cooling parts of the curve separately and independently [8]. For this reason, the time needed for the analysis of thermal diffusivity via the curve-fitting method is in the

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order of seconds. Owing to this long analysis time, the measured thermal diffusivity could also be influenced by external factors other than the radiative heat loss, such as the signal stability of the temperature-response curve. Therefore, it is necessary to develop a suitable method for analyzing thermal diffusion in solid materials at high temperature, which does not require measurement over a long temperature-response period.

The logarithmic method was suggested by Takahashi et al., Azumi, and Thermitus and Laurent [9–11]. This method utilizes Laplace transformation and term-by-term inversion, and the derived equation for the temperature-response curve shows good convergence at the early stage of the temperature rise on the rear surface. It has been shown in [9–11] that the logarithmic method is insensitive to nonideal conditions and does not require correction procedures. Moreover, the logarithmic method has the advantage of rapidity – less than 1 s is required for analysis.

In this study, considering the abovementioned advantages of the logarithmic method, we analyzed the effect of radiative heat loss in the application of this method. In addition, simulations, using the thermal diffusivity data on copper (Cu), iron (Fe), tungsten (W), alumina–titanium carbide ceramics ($\text{Al}_2\text{O}_3\text{--TiC}$), and aluminum silicate (AS), available in the literature, were used to estimate the usefulness of the normalized logarithmic method. AS was used as an example of a low-thermal-diffusivity sample.

2 Theory

2.1 Theoretical temperature-response curves with radiative heat loss

When radiative heat loss becomes significant, the temperature on the rear surface of a sample reaches its maximum and decreases according to the equations given in [4,5,12]:

$$\frac{T}{T_M} = \sum_{n=0}^{\infty} A_n \exp[-X_n t^+], \quad (1)$$

where

$$A_n = 2(-1)^n X_n^2 (X_n^2 + 2Y + Y^2)^{-1} \quad (2)$$

and

$$X_0 = (2Y)^{0.5} \left(1 - \frac{Y}{12} + \frac{11Y^2}{1,440} \right). \quad (3)$$

For $n \geq 1$,

$$X_n = n\pi + \frac{2}{n\pi} Y - \frac{4}{(n\pi)^3} Y^2 + \left(\frac{16}{(n\pi)^5} - \frac{2}{3(n\pi)^3} \right) Y^3 + \left(\frac{-80}{(n\pi)^7} + \frac{16}{3(n\pi)^5} \right) Y^4, \quad (4)$$

where T_M , t^+ , and Y are the maximum temperature rise under adiabatic conditions, the dimensionless elapsed time (the Fourier number), and the Biot number, respectively. The latter two parameters are defined as follows:

$$t^+ = \frac{\alpha t}{l^2}, \quad Y = \frac{4\varepsilon\sigma T^3 l}{\lambda}, \quad (5)$$

where l is the thickness of the sample, α is the thermal diffusivity of the sample, t is the elapsed time, λ is the thermal conductivity of the sample, ε is the thermal emissivity of the sample surface, σ is the Stefan–Boltzmann constant, and T is the temperature of the sample. The temperature-response curve was normalized using the maximum temperature rise, T'_M . The normalized temperature rise is defined as T/T'_M . This means that the normalized temperature-response curves can only be discussed by using the dimensionless elapsed time and the Biot number. The normalized temperature-response curve is plotted in Figure 1. Compared to the situation under adiabatic conditions, the theoretical temperature with radiative heat loss reaches the maximum value of T/T'_M earlier and decreases after reaching the maximum value.

2.2 Principle of logarithmic method

In the conventional halftime method, the ideal one-dimensional thermal diffusion equation without

Figure 1: Normalized theoretical temperature-response curves considering radiative heat loss.

radiative heat loss is used. The normalized temperature rise at the rear surface of the sample, T/T'_M , is given by the following Fourier series [3]:

$$T/T'_M = 1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp\left(\frac{-n^2 \pi^2 \alpha t}{l^2}\right) \quad (6)$$

Further, by solving the one-dimensional thermal diffusion equation using Laplace transformation and term-by-term inversion, the following equation can be obtained:

$$T/T'_M = \frac{2l}{(\pi \alpha t)^{0.5}} \sum_{n=1}^{\infty} \exp\left(-\frac{(2n-1)^2 l^2}{4 \alpha t}\right) \quad (7)$$

Equation (7) shows very good convergence at the early stages of the elapsed time, and T/T'_M can be calculated using one term over the region with $T/T'_M = 0.9$ as follows:

$$T/T'_M = \frac{2l}{(\pi \alpha t)^{0.5}} \exp\left(-\frac{l^2}{4 \alpha t}\right) \quad (8)$$

Equation (8) is transformed and logarithmized as follows:

$$\ln(t^{0.5} T/T'_M) = \ln\left(2 \left(\frac{l^2}{\pi \alpha}\right)^{0.5}\right) - \frac{l^2}{4 \alpha t} \quad (9)$$

When the thermal diffusivity is analyzed at high temperatures using the normalized logarithmic method, it is determined using the following equation:

$$\ln(t^{0.5} T) = \ln\left(2 T'_M \left(\frac{l^2}{\pi \alpha}\right)^{0.5}\right) - \frac{l^2}{4 \alpha t} \quad (10)$$

As the relationship between $\ln(t^{0.5} T)$ and $\frac{1}{t}$ is linear, thermal diffusivity is obtained from the slope, $-\frac{l^2}{4 \alpha}$, of the plot as a function of $\frac{1}{t}$.

Considering the good convergence shown at the early stages of the elapsed time and during the laser irradiation time, the best T/T'_M range is generally from 0.3 to 0.6 [8]. Thus, in this study, the effect of radiative heat loss was analyzed using the logarithmic method in the T/T'_M range of 0.3–0.6.

Equation (8) can be written using dimensionless parameters as follows, with $t^+ = \frac{\alpha t}{l^2}$:

$$T/T'_M = \frac{2}{(\pi t^+)^{0.5}} \exp\left(-\frac{1}{4 t^+}\right). \quad (11)$$

Equation (11) is transformed and logarithmized as follows:

$$\ln((t^+)^{0.5} T/T'_M) = \ln(2 \pi^{-0.5}) - \frac{1}{4 t^+} \quad (12)$$

To evaluate the effect of radiative heat loss from the solid sample on thermal diffusivity, the relationship

Figure 2: Normalized theoretical temperature-response curves in the T/T'_M range from 0.3 to 0.6: (a) plot of $f = \ln((t^+)^{0.5} T/T'_M)$ versus $1/t^+$ and (b) slope $k = \frac{df}{d(1/t^+)}$ versus $1/t^+$.

between $f = \ln((t^+)^{0.5} T/T'_M)$ and is examined. We define the slope of f versus $1/t^+$ as $k = \frac{df}{d(1/t^+)}$. In case of the adiabatic conditions, $k = -1/4 (= -0.25)$.

3 Simulation – results and discussion

The normalized theoretical temperature-response curves in the T/T'_M range of 0.3–0.6, which were calculated using equation (1), are shown in Figure 2. The $1/t^+$ dependence of f is shown in Figure 2(a), and the $1/t^+$ dependence of the slope $k = \frac{df}{d(1/t^+)}$ is shown in Figure 2(b).

Figure 3: Biot number dependence of slope $k = \frac{df}{d(1/t^+)}$ and its deviation $De(\%)$.

Table 1: Calculated Biot number Y , deviation of slope De (%), input thermal diffusivity values α' ($\text{m}^2 \text{s}^{-1}$), estimated thermal diffusivity values α ($\text{m}^2 \text{s}^{-1}$), and thermal diffusivity values evaluated by halftime method α_h ($\text{m}^2 \text{s}^{-1}$) of copper (Cu), tungsten (W), iron (Fe), alumina–titanium carbide ceramics (Al_2O_3 –TiC), and aluminum silicate (AS) samples at various temperatures T (K)

Sample	T (K)	Y (10^{-5})	De (%)	α' ($\text{m}^2 \text{s}^{-1}$)	α ($\text{m}^2 \text{s}^{-1}$)	α_h ($\text{m}^2 \text{s}^{-1}$)
Cu thickness: 4 mm	300	5.49	−0.001	1.17×10^{-4}	1.17×10^{-4}	1.17×10^{-4}
	600	46.5	−0.008	1.04×10^{-4}	1.04×10^{-4}	1.04×10^{-4}
	800	114	−0.020	9.77×10^{-5}	9.77×10^{-5}	9.78×10^{-5}
	1,000	231	−0.041	9.16×10^{-5}	9.16×10^{-5}	9.19×10^{-5}
W thickness: 1 mm	300	1.06	0.000	6.83×10^{-5}	6.83×10^{-5}	6.83×10^{-5}
	600	10.7	−0.002	5.09×10^{-5}	5.09×10^{-5}	5.09×10^{-5}
	800	27.9	−0.005	4.50×10^{-5}	4.50×10^{-5}	4.50×10^{-5}
	1,000	576	−0.010	4.12×10^{-5}	4.12×10^{-5}	4.12×10^{-5}
Fe thickness: 1 mm	300	1.98	0.000	2.61×10^{-5}	2.61×10^{-5}	2.61×10^{-5}
	600	14.7	−0.003	2.25×10^{-5}	2.25×10^{-5}	2.25×10^{-5}
	800	54.0	−0.009	1.24×10^{-5}	1.24×10^{-5}	1.24×10^{-5}
	1,000	110	−0.019	8.30×10^{-6}	8.30×10^{-6}	8.31×10^{-6}
Al_2O_3 –TiC thickness: 1 mm	300	30.6	−0.005	9.51×10^{-6}	9.51×10^{-6}	9.51×10^{-6}
	600	245	−0.043	3.65×10^{-6}	3.65×10^{-6}	3.66×10^{-6}
	800	581	−0.102	2.95×10^{-6}	2.95×10^{-6}	2.97×10^{-6}
	1,000	1,130	−0.198	2.60×10^{-6}	2.60×10^{-6}	2.64×10^{-6}
AS thickness: 1 mm	373	50.0	−0.009	1.714×10^{-6}	1.713×10^{-6}	1.714×10^{-6}
	1,273	2,010	−0.348	1.047×10^{-6}	1.043×10^{-6}	1.068×10^{-6}

In Figure 2(a), knowing that the intercept is fixed, it is evident that the slope gently shifts as the Biot number (Y) increases. On the other hand, in Figure 2(b), the slope is $-1/4$ ($=-0.250$), as shown in equation (12), under adiabatic conditions; but k gradually shifts with increasing Biot number because of radiative heat loss. Thus, the deviation of the slope corresponds to the effect of radiative heat loss on thermal diffusivity. The deviation of k , De (%), is given as follows:

$$De(\%) = 100 \times \frac{(-0.250) - k}{-0.250} \quad (13)$$

In addition, it was also found that the shift in k at early times (at larger values of $1/t^+$) is smaller than that at later times (at smaller values of $1/t^+$). This means that the effect of radiative heat loss on temperature response during the earlier stage of the elapsed time is smaller than that during the later stage of the elapsed time.

The dependence of k and De on the Biot number is shown in Figure 3. We observed De at $Y = 0.1$ to be approximately -1.6% and that at $Y = 0.2$ to be approximately -3.0% . Thus, when the temperature-response curve can be measured up to the elapsed time at T'_M , the obtained thermal diffusivity of the solid sample with a low thermal diffusivity, at $Y = 0.1$, using the normalized logarithmic method, is lower by approximately 1.6% than the input thermal diffusivity values.

In Figure 3, De is expressed as a function of Y in the following manner:

$$De(\%) = 0.125Y^2 - 0.176Y \quad (15)$$

Table 1 presents the Y values, calculated using equation (5), $De(\%)$ values, input thermal diffusivity values α , estimated thermal diffusivity values α' , and thermal diffusivity evaluated by the halftime method α_h ; each of these values is listed for Cu, W, Fe, Al_2O_3 –TiC, and AS samples at various temperatures. For the evaluation of this data, the thickness of the sample, l , was assumed to be 4 mm for Cu and 1 mm each for W, Fe, Al_2O_3 –TiC, and AS. The estimated α values of Cu, W, Fe, Al_2O_3 –TiC, and AS using equation (14) were obtained from literature [13–16]. The thermal emissivity of the sample surface, ε , was assumed to be 0.9 for Cu, Al_2O_3 –TiC, and AS because graphite powder was sprayed on the front and rear surfaces of the Cu and AS samples before the thermal diffusivity measurements were performed; the ε value of graphite is approximately 0.9. However, the ε values of the W and Fe samples were both assumed to be 0.3 because graphite powder was not sprayed on these samples. The λ values of Cu, W, Fe, Al_2O_3 –TiC, and AS were also obtained from literature [13–16]. The results show that when AS was heated to 1,273 K, the maximum deviation of the obtained thermal diffusivity from the input value was approximately -0.35% . In contrast, the difference between the input value ($1.047 \times 10^{-6} \text{m}^2 \text{s}^{-1}$) and the thermal diffusivity evaluated by the halftime method ($1.068 \times 10^{-6} \text{m}^2 \text{s}^{-1}$) when AS was heated to

1,273 K was approximately 2.38%. Therefore, since we observed that the effect of radiative heat loss on thermal diffusivity at high temperature was negligibly small, we conclude that the normalized logarithmic method is very useful for analyzing the thermal diffusivity of low-thermal-diffusivity solid samples.

4 Conclusions

We utilized the logarithmic method to evaluate the thermal diffusivity of low-thermal-diffusivity solid samples. The logarithmic method can determine the thermal diffusivity during the early stages of the response time. In this study, we found that the effect on the thermal diffusivity of radiative heat loss from the solid sample could be evaluated using the normalized logarithmic method at high temperatures. When the Biot number was 0.1, the deviation of the obtained thermal diffusivity was approximately –1.6% from the input value. When AS was heated to 1,273 K, the maximum deviation was approximately –0.35%. In contrast, the difference between the input value ($1.047 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$) and the thermal diffusivity evaluated by the halftime method ($1.068 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$) when AS was heated to 1,273 K was approximately 2.38%. Therefore, as the effect of radiative heat loss on thermal diffusivity at high temperatures was found to be negligibly small, the normalized logarithmic method, which has the advantage of a short analysis time (less than 1 s), is deemed to be very useful for the analysis of the thermal diffusivity of solid samples with low thermal diffusivity.

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References

- [1] Li, M., and M. Akoshima. Appropriate metallic coating for thermal diffusivity measurement of nonopaque materials with laser flash method and its effect. *International Journal of Heat and Mass Transfer*, Vol. 148, 2020, p. 119017.
- [2] Nishi, T., M. Takano, K. Ichise, M. Akabori, and Y. Arai. Thermal conductivities of Zr-based transuranium nitride solid solutions. *Journal of Nuclear Science and Technology*, Vol. 48, 2011, pp. 359–365.
- [3] Parker, W. J., R. J. Jenkins, C. P. Butler, and G. L. Abbott. Flash method of determining thermal diffusivity, heat capacity, and thermal conductivity. *Journal of Applied Physics*, Vol. 32, 1961, pp. 1679–1684.
- [4] Cezairliyan, A., T. Baba, and R. Taylor. A high-temperature laser-pulse thermal diffusivity apparatus. *International Journal of Thermophysics*, Vol. 15, 1994, pp. 317–341.
- [5] Josell, D., J. Warren, and A. Cezairliyan. Comment on “Analysis for determining thermal diffusivity from thermal pulse experiments.” *Journal of Applied Physics*, Vol. 78, 1995, pp. 6867–6869.
- [6] Akoshima, M., and T. Baba. Thermal diffusivity measurements of candidate reference materials by the laser flash method. *International Journal of Thermophysics*, Vol. 26, 2005, pp. 151–163.
- [7] Nishi, T., M. Takano, A. Itoh, M. Akabori, Y. Arai, K. Minato, et al. Thermal conductivity of AmO_{2-x} . *Nuclear Materials*, Vol. 373, 2008, pp. 295–298.
- [8] Baba, T., and A. Ono. Improvement of the laser flash method to reduce uncertainty in thermal diffusivity measurements. *Measurement Science and Technology*, Vol. 12, 2001, pp. 2046–2057.
- [9] Takahashi, Y., K. Yamamoto, and T. Ohsato. Advantages of logarithmic method – a new method for determining thermal diffusivity- in the laser-flash technique. *Netsu Sokutei*, Vol. 15, 1988, pp. 103–109 (in Japanese).
- [10] Azumi, T., Measurement technology of Laser flash method. *Metal & Technology*, Vol. 9, 1992, pp. 1–9 (in Japanese).
- [11] Thermitus, M.-A., and M. Laurent. New logarithmic technique in the flash method. *International Journal of Heat and Mass Transfer*, Vol. 40, 1997, pp. 4183–4190.
- [12] Cape, J. A., and G. W. Lehman. Temperature and finite pulse-time effects in the flash method for measuring thermal diffusivity. *Journal of Applied Physics*, Vol. 34, 1963, pp. 1909–1913.
- [13] Shinpen Netsu Bussei Handbook, edited by the Japan Society of Calorimetry and Thermal Analysis (Yokendo, Tokyo, 2008) p. 23, Vol. 213, 276 (in Japanese).
- [14] Reference Material Certificate of NMIJ CRM 5807-a for Al_2O_3 -TiC Ceramics for Thermal Diffusivity Measurement. https://unit.aist.go.jp/nmij/english/refmate/crm/certificate_sds/90/5807a_en.pdf.
- [15] Yamada, H., T. Ishii, and H. Hara. New Ceramics Tools. *Hitachi Review*, Vol. 64, 1982, pp. 63–68 (in Japanese).
- [16] Metal Databook, 4th ed., edited by the Japan Institute of Metals and Materials (Maruzen, Tokyo, 2004) p. 90 (in Japanese).