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Thermal Diffusivity Measurement of Porous Materials†

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Abstract

The improved method to measure the thermal diffusivity of porous materials such as ceramics is proposed in using an ordinary laser flash method. The penetration of the laser energy into the test piece of porous materials is taken account in this method, by using the derived theoretical equation. The thermal diffusivity of such materials and its measurement errors are calculated simultaneously by fitting the experimental data to this derived equation with a personal computer connected to the laser flash device. The usefulness of this method is shown by comparing it with the conventional method.

KEY WORDS: (Thermal Diffusivity) (Porous Materials) (Laser Flash Method) (On-Line Dialogue Type Measurement)

1. Introduction

Thermal conductivity, specific heat and thermal diffusivity are important quantities that prescribe the characteristics of heat transfer of materials. The accurate measurement of these values is indispensable in scientific and engineering fields.

Usually this measurement is achieved by heating a test piece and then estimating the hypothetical values from the variation in temperature by using theoretical equations. Various methods are available for heating the test piece and each has its own merits and demerits[1].

The laser flash method, a type of pulse heating method, is widely used as a relatively easy method. However, it has some disadvantages. For example, when evaluating thermal diffusivity in ceramics by the laser flash method, if laser energy penetrates into the test piece, the application of conventional theories might lead to considerable error.

In this research, applying the laser flash method to penetrative materials, theoretical equations were derived for taking into account laser energy penetration into the test piece, a method of accurately measuring thermal diffusivity for practical application was found, and thermal diffusivity was measured and measurement errors were evaluated.

2. Theory of Calculation of Thermal Diffusivity

According to the conventional method, thermal diffusivity a is measured using Eq.(1)[2].

$$a = 1.37 d^2 / \pi^2 t_{0.5} \tag{1}$$

In Eq.(1), d is the thickness of the test piece as shown in **Fig. 1,** and t is the time required for the temperature rise curve of the observed surface, the back side, of the test piece to reach half of its maximum value (refer to **Fig. 2**) after the laser flash and the heat input to the front side. Equation (1) can be obtained from Eq.(2) where temperature rise T on the back side has been calculated with the
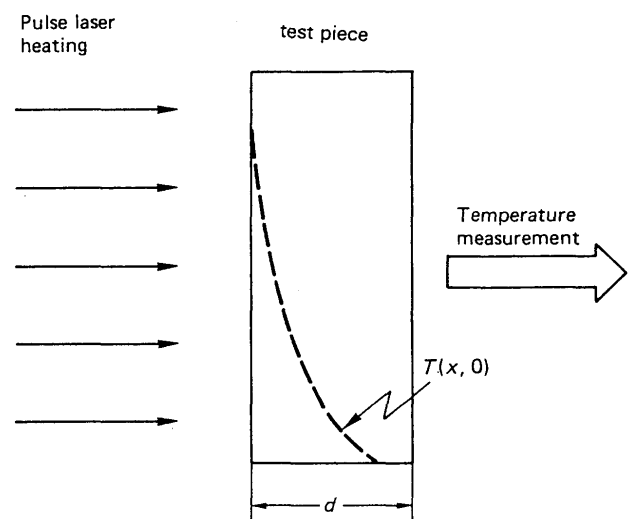

Fig. 1 Schematics of thermal diffusivity measurement by laser flashing
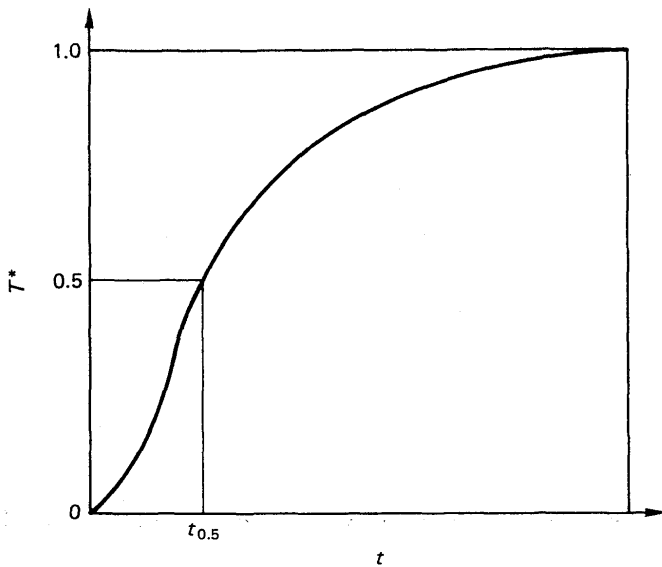
Fig. 2 Temperature rise pattern of test piece during measurement and definition of $t_{0.5}$

Fig. 3 Temperature rising theoretically calculated by taking into account for penetration of laser flash energy

one-dimensional heat conduction theory of an infinite slab of finite thickness when heat input is applied to the surface of the test piece also shown in Fig. 1 by the flash of the laser. The effect of thermal radiation can be neglected in this equation provided the temperature of the test piece is below 1000°C.[3]

$$T^* = 1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp(-n^2 \pi^2 t^*) \qquad (2\text{-}1)$$

$$T^* = cd\rho T / I_0 \qquad (2\text{-}2)$$

$$t^* = at / d^2 \qquad (2\text{-}3)$$

where c: the specific heat, ρ: the density of the test piece, and I_0: the input energy of the incident laser beam. Dimensionless variables of temperature and time are expressed by attaching a superscript asterisk. In Eq.(2–1), by setting $T^* = 0.5$, Eq.(1) can be obtained by the numerical calculation with $t^* = 0.5$. In Eq.(2), as the input laser energy is regarded as heating only the surface of the test piece, penetration inside is not considered.

When penetration of energy into the test piece is considered, the initial temperature distribution within the test piece should be expressed by Eq.(3) due to the penetration of energy when the laser is applied.

$$T(x,0) = \mu I_0 \exp(-\mu x) / c\rho \qquad (3)$$

Here μ is the absorption coefficient of laser energy, which is the quantity to express the penetration into the test piece. With Equation (3) employed as the initial condition, if the equation of one-dimensional heat condition is solved by the same boundary condition as in the case where Eq. (2) was obtained, then Eq.(4) can be obtained.
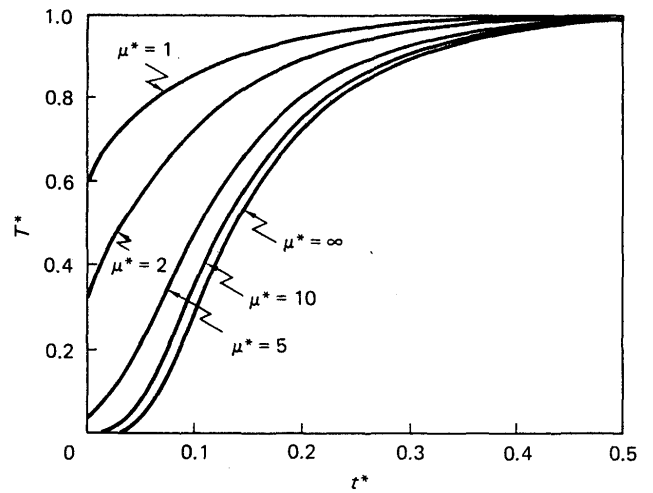
$$T^*(d,t) = 1 + \frac{2}{[1 - exp(-\mu^*)]} \cdot$$

$$\sum_{n=1}^{\infty} [(-1)^n - \exp(-\mu^*)] \frac{\exp(-n^2 \pi^2 t^*)}{[1 + (n\pi/\mu^*)^2]} \qquad (4)$$

Here, $\mu^* = \mu d$ (dimensionless absorption coefficient), and when $\mu - > \infty$ (there is no penetration), Eq.(4) becomes Eq.(2-1). With μ^* as the parameter, **Fig. 3** can be obtained by the numerical calculation of Eq.(4). As seen from the figure, if μ^* is small and the penetability great, then the temperature rises rapidly, of course. In this case, the value for thermal diffusivity may be too high if penetability is not taken into account. In order to evaluate the error quantitatively at this case, $_\mu t^*_{0.5}$ is obtained from Eq.(4) by numerical calculation for each value of μ^*, and calculation is done with the difference of $t^*_{0.5}$ obtained from Eq.(2) regarded as the relative value Δt^* (Eq.(5)).

$$\Delta t^* = (_\mu t^*_{0.5} - t^*_{0.5}) / _\mu t^*_{0.5} \qquad (5)$$

Figure 4 shows the graph thus obtained. From the definition of Fourier's Number, it is obvious that this Δt^* corresponds to the relative error that occurs when obtaining thermal diffusivity from Eq.(2) while the penetration of laser energy into the test piece is present. From Fig. 4, it is understood that this error cannot be ignored when the dimensionless absorption coefficient is less than 15 or so, particularly when less than 5. By increasing the thickness of the test piece, this error may be reduced as μ^* is enlarged. However, measurement becomes less accurate then because the thicker the test piece, the smaller the temperature increase on the observed surface. Hereinafter the evaluation of thermal diffusivity a and absorption coefficient μ^* resulting from the test data and Eq.(4) will be examined.
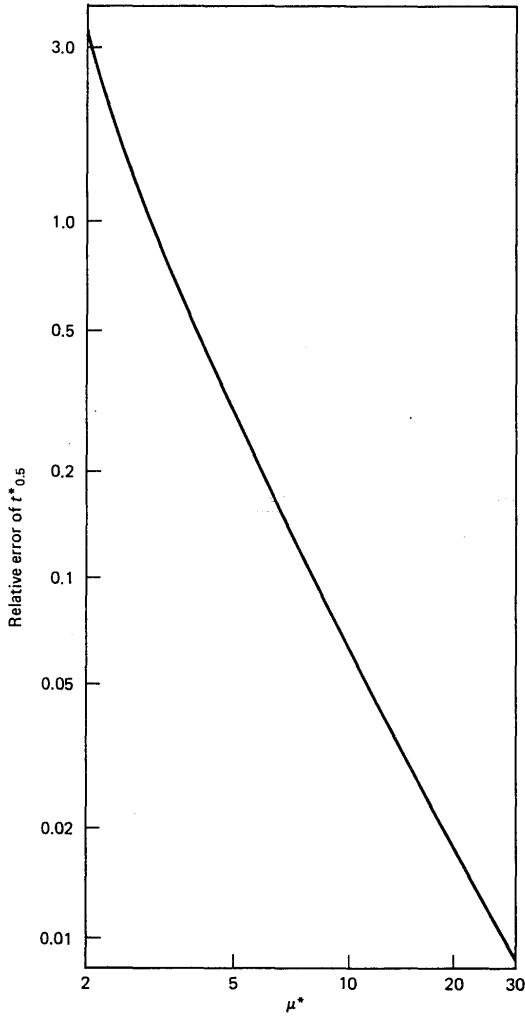
Fig. 4 Error of thermal diffusivity value estimated in case of neglecting penetration effect of laser flash energy

Fig. 5 Flowchart of experimental data processing for calculation of thermal diffusivity and absorption coefficient

3. Calculation of Absorption Coefficient and Thermal Diffusivity

3.1 Application of observation equation and least squares method

More than two simultaneous equations are necessary to find μ^* and a from Eq.(4). Therefore, N sets of (t_i, T_i) $(i = 1 \ldots \ldots N)$ (hereinafter the superscript asterisk is omitted for convenience of reading the equations) are found from such temperature rise curves, as shown in Fig. 2, obtained by experiment. Substituting these values in Eq. (4) yield N sets of observation equations, Eq(6).

$$T_i = F_{0i}(a,\mu,t_i) \qquad i = 1,2,\ldots,N \qquad (6)$$

By linearizing Eq.(6) with the approximate values of μ and a supposed to be μ_0 and a_0 respectively, the normal equation was derived from the principle of the least squares method.
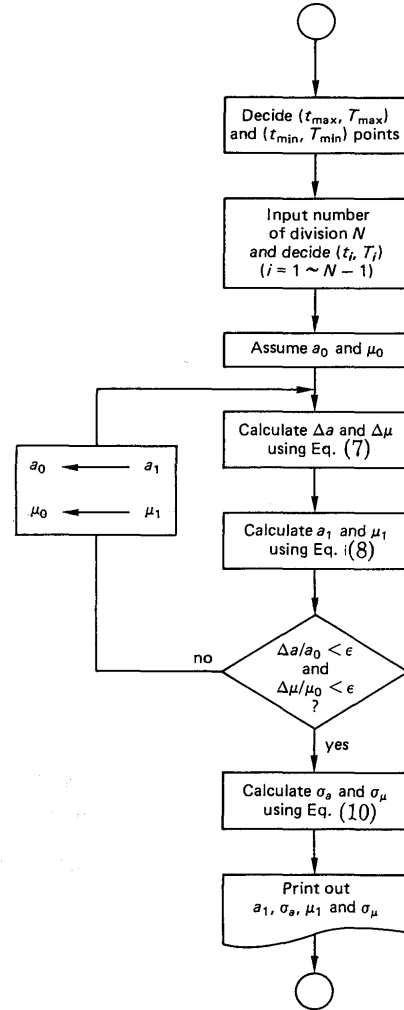
$$\Delta\mu \sum_{i=1}^{N} A_i^2 + \Delta a \sum_{i=1}^{N} A_i B_i = \sum_{i=1}^{N} \Delta T_i A_i \ \Bigg\}$$

$$\Delta\mu \sum_{i=1}^{N} A_i B_i + \Delta a \sum_{i=1}^{N} B_i^2 = \sum_{i=1}^{N} \Delta T_i B_i \ \Bigg\} \qquad (7)$$

$$A_i = \partial F_{0i}/\partial \mu_0 \ \Bigg\}$$
$$B_i = \partial F_{0i}/\partial a_0$$
$$\Delta T_i = F_{0i}(a_0, \mu_0, t_i) - T_i$$

Solving Eq.(7), and we can find $\Delta\mu$ and Δa, and corrections for approximate values by using Eq.(8) yields the most probable values μ_1, a_1.

$$\mu_1 = \mu_0 - \Delta\mu \ \Bigg\}$$
$$a_1 = a_0 - \Delta a \ \Bigg\} \qquad (8)$$

Further, since the residual equation can be expressed by Eq.(9),

$$\Delta T_{1i} = T_i - F_{0i}(a_i,\mu_1,t_i) \qquad (9)$$

the estimated standard deviations of the most probable

values μ_1, and a_1 can be calculated by

$$\sigma_\mu = \left\{ \sum_{i=1}^{N} \Delta T_i \sum_{i=1}^{N} B_i^2 \Big/ (N-2) \right.$$

$$\left. \left[\sum_{i=1}^{N} A_i^2 \sum_{i=1}^{N} B_i^2 - \left(\sum_{i=1}^{N} A_i B_i \right)^2 \right] \right\}^{1/2} \qquad (10)$$

$$\sigma_a = \sigma_\mu \left\{ \sum_{i=1}^{N} A_i^2 \Big/ \sum_{i=1}^{N} B_i^2 \right\}^{1/2}$$

3.2 Calculation by means of personal computer

The use of a personal computer facilitates the evaluation of μ and a by applying Eqs.(7)~(10) to the experimental results. First, convert the analogue output signal from the laser flash device to A/D, and input it into a computer along with sampling frequency and test piece thickness. Next, processing the input data according to the flowchart

(a)

(c)

(b)
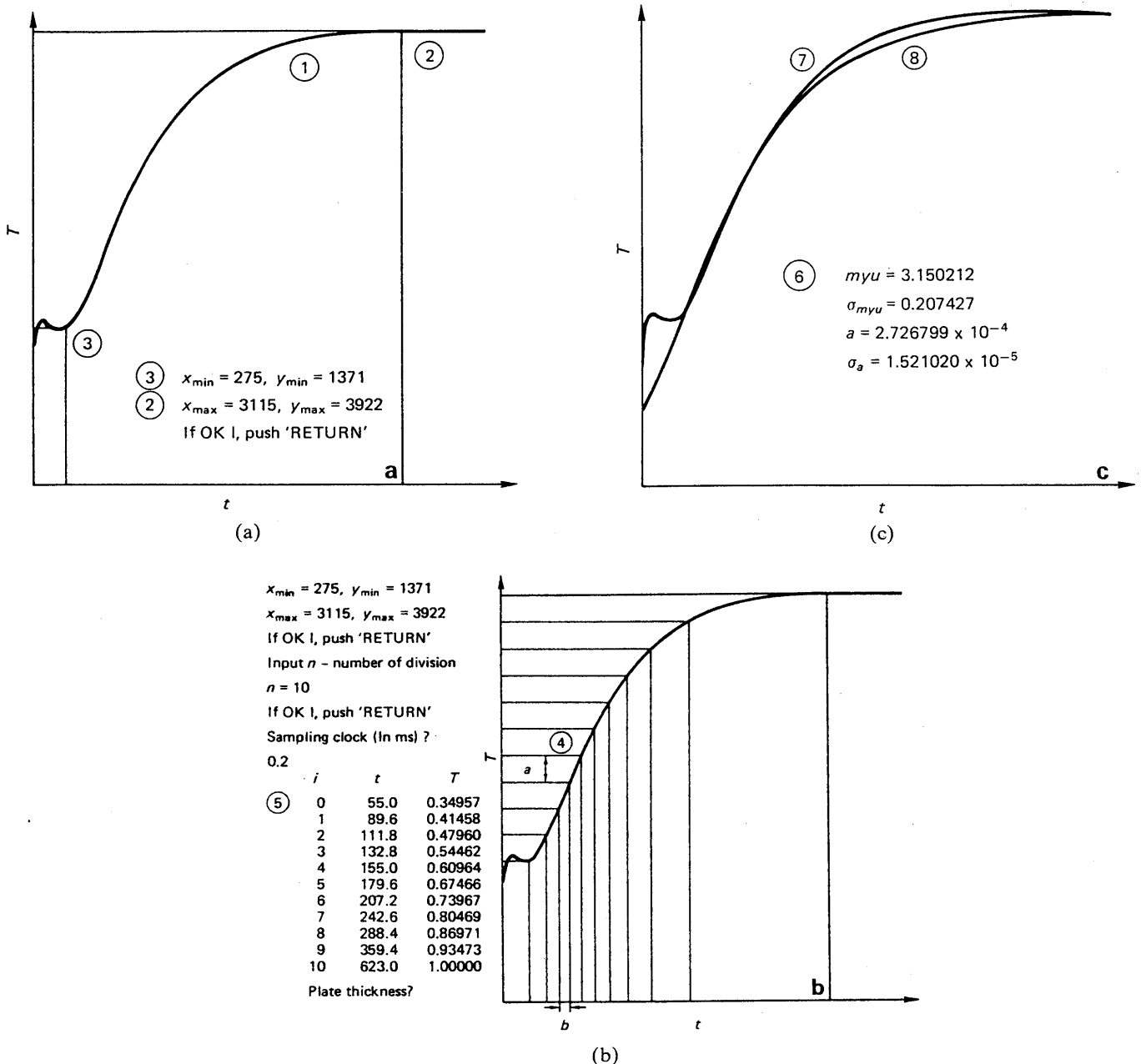
Fig. 6 Example of measurement
(a) Determination of T_{max} and T_{min} points on experimentally measured temperature rise curve
(b) Sampling data set (t_i, T_i) $(i=0...10)$ between T_{max} and T_{min}
(c) Comparison of original and derived data, most probable values were substituted in to the latter

shown in **Fig. 5** as follows. At the first step, plot the data on a time axis. Find their maximum value (t_{max}, T_{max}) and indicate it as the graph of the display of the computer. The position (t_{min}, T_{min}) of the minimum value (regarded as unaffected by laser flash) of the data can be specified manually by the operator by moving the cursor along the data graph. At the next step, input the number of samplings N, divide the distance between T_{max} and T_{min} by N, compute T_i($T_0 = T_{min}$, $i = 0$ ... ... $N - 1$, T_{max} should not be used), and search the data to find t_i corresponding to each T_i. When searching the data, we must be careful to minimize the influence of noise overlapping on the data.

With respect to the N sets of (t_i, T_i), calculate the correction values $\Delta\mu$, Δa of each approximate value according to Eq.(7). To make this calculation, assume the initial approximate values μ_0 and a_0 suitably, modify the approximate values by the computed correction values, repeat the calculation until the relative values of the correction values become less than the allowable limit using the modified values as the next approximate values ($\mu_1 \dashrightarrow > \mu_0$, $a_1 \dashrightarrow > a_0$), and let the values modified by these correction values be the most probable. In this calculation loop, if μ_1 becomes greater than a certain set value, calculate a_1 alone by Eq.(11), based on Eq.(2), instead of by Eq.(7).

$$\left. \begin{array}{l} \Delta a = \sum_{i=1}^{N} \Delta T_i B_i / \sum_{i=1}^{N} B_i^2 \\ a_1 = a_0 - \Delta a \end{array} \right\} \qquad (11)$$

Finally, according to Eq.(10) or (12) (the following equation) calculate estimated standard deviation for the most probable value as

$$\sigma_a = \left\{ \sum_{i=1}^{N} \Delta T_{1i}^2 / (N-1) \sum_{i=1}^{N} B_i^2 \right\}^{1/2} \qquad (12)$$

The calculation described is composed in such a manner that it is achieved as a dialogue with the computer while the operator confirms the processing results at each step watching graphs or numerical values.

3.3 Example of measurement

With reference to **Fig. 6,** an example will be described in which the thermal diffusivity of sintered ceramics $Z_rO_2 - 3mol\%Y_2O_3$ cut to the thickness of 0.63 mm was observed. Figure 6 (a) shows the result of setting the maximum value position ② and the minimum value position ③ by digitally converting the temperature rise signals due to lase flashing (energy : 6J, wavelength : 694.3 nm, pulse width : 0.7 mS) into 12 bits and 4 KW and plotting them at $t - T$ coordinates as position ① in the aforementioned manner.

When this result was accepted as reasonable, the required number N of sampling data ($N = 10$ in this example) was input, then each partition point (t_i, T_i) ($i =$

Table 1 Comparison of present method with conventional method in thermal diffusivity measurement of $ZrO_2 - 3mol\%Y_2O_3$

	Present Method	Conventional Method
Thermal Diffusivity a(m²/s)	2.91×10^{-7}	4.66×10^{-7}
Stand. Deviation σ_a(m²/s)	9.88×10^{-9}	
RMS of Residuals	1.47×10^{-1}	8.71×10^{-1}

$0 ... N$) was indicated by graph (Fig. 6 (b), position ④) and numerical values ⑤. When that also was accepted, iterative approximation by Eq.(8) was made (for about 1 sec. on an ordinary personal computer with 16-bit numerical data processor) to calculate the most probable values of μ and a. Then the estimated values of standard deviation of these most probable values were calculated by Eq.(10) and output to the printer along with the most probable values as shown ⑥ in Fig.6 (c). If required, the original data ⑦ can be compared with the date ⑧, obtained by substituting these most probable values in Eq.(4), by plotting on the graph.

3.4 Comparison with conventional method and supplementary explanation

In the example above, thermal diffusivity was calculated according to Eq.(8). **Table 1** shows a comparison of the example with the results from using the conventional method of Eq.(1). As shown in the table, the thermal diffusivity increases about 60% when calculated by the latter. This difference almost agrees with the value estimated from the calculated absorption coefficient and Fig. 4. Furthermore, by the conventional method, if the estimated standard deviation of the evaluate value is calculated from Eq.(10) and so on, then it approaches infinity (due to the use of experimental value obtained at one point only).

The RMS of residuals in this table was calculated from the difference between the experimental data and the theoretical values by using the computed most probable values from Eq. (13), and serves as a measure of fidelity of the two methods to the experimental values. As shown in the table, this value for the conventional method is about six times the values for the present method

$$\sigma = \left\{ \sum_{i=1}^{N} \Delta T_{1i}^2 / N \right\}^{1/2} \qquad (13)$$

Table 2 Comparison of present method with conventional method in thermal diffusivity measurement of ordinary metal (carbon steel S45C)

Thermal Diffusivity a(m²/s)	Present Method	Conventional Method
Thermal Diffusivity a(m²/s)	1.11×10^{-1}	1.09×10^{-1}
Stand. Deviation σ_a(m²/s)	1.64×10^{-7}	
RMS of Residuals	2.35×10^{-1}	2.54×10^{-1}

When penetration of laser energy into the test piece need not be considered, for example, if applying Eq.(8) to a metal (carbon steel S45C) test piece, the iterative approximation is carried out, the absorption coefficient μ becomes greater, and as mentioned above, the calculation enters the routine according to Eq.(11), resulting in the calculation of thermal diffusivity a alone. In **Table 2,** as in Table 1, this value is shown along with the value obtained by the conventional method and respective values of the RMS of residuals resulting from Eq. (13). Since the difference between the RMS of residuals made by the two methods is relatively small, the data at $t_{0.5}$ point represents fairly well the characteristics of the rise in temperature of the test piece when there is no penetration. Yet this method is higher in fidelity.

With the previously mentioned matter of the calculation of standard deviation estimate also taken into account, this method can bring about better results, even when the penetration into the material need not be considered in particular.

4.   Conclusion

The rsults of this study are summarized as follows.
1) The propose is made that when thermal diffusivity is evaluated by the application of the laser flash method to a material into which laser energy penetrates, Eq.(4) which considers the penetration of energy into the material as its absorption coefficient should be used.

2) The error found when the conventional method is applied to penetrative materials has been estimated, and verification has been made that errors larger than 20% may often generate if the penetration is to a great extent.
3) By the least squares method, equations for the evaluation of absorption coefficient, thermal diffusivity and estimates of their standard deviation have been derived (Eq. (8), (9), (10), (11)) from the experimental data and Eq.(4), and examples have been shown of calculation in an on-line measurement system interfacing the laser flash device to a personal computer.
4) This method has been applied to ceramics and metals, and thermal diffusivities evaluated, then compared with the conventional method to reveal its usefulness and feasibility.

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