Study on a thermal diffusivity standard for the laser flash method measurements[1]

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Abstract

The National Metrology Institute of Japan (NMIJ) in AIST has been studying the laser flash method in order to establish the SI traceable thermal diffusivity standard. We have developed key technologies to reduce uncertainty in laser flash measurements. This time we carried out an uncertainty evaluation on the laser flash measurement in order to determine the thermal diffusivity value of IG-110, a grade of isotropic high-density graphite, as a candidate reference material. Thermal diffusivity on the laser flash method is derived quantity from a specimen thickness and a thermal diffuse time. And thermal diffusivity values of materials are a function of temperature. The measurement system is also composed of three units corresponding to each quantities, length, time, and temperature. Then we checked and calibrate our measurement system and estimated the uncertainty of a measurement result.

KEY WORDS: laser flash method; uncertainty; reference material; solid material; thermal diffusivity

1. INTRODUCTION

The flash method is one of the most popular methods to measure thermal diffusivity of solid materials above room temperature [1]. The National Metrology Institute of Japan (NMIJ) in AIST has been studying the laser flash method in order to establish a reference material as a thermal diffusivity standard [2,3]. We hope that a reference material is useful to check the verification of laser flash instruments. We have developed key technologies to reduce uncertainty in laser flash measurements [2]. For example, a uniform pulse heating, development of a fast infrared radiation thermometer, introduction of a new data analysis algorithm, and an extrapolated method to determine an intrinsic thermal diffusivity value. We have also investigated candidate reference materials for laser flash measurement based on following concepts:

1) It has good homogeneity and stability.
2) It can be measured without black coatings.
3) It is a set of some specimens from a same substance with different thickness.
4) Uncertainty of a thermal diffusivity value is evaluated.
5) The thermal diffusivity value of a set is SI traceable value.

From our research, it is found that IG-110 is appropriate for a reference material for laser flash measurements. IG-110, a grade of isotropic graphite manufactured by Toyo Tanso Co., Ltd, is black. It showed good homogeneity and stability [3,4]. From measurements changing pulse-heating energy, it is confirmed that the thermal diffusivity values of different thickness IG-110 specimens from one lot agreed with each other within their homogeneity. Then we carried out an uncertainty evaluation on the laser flash measurement in order to determine the SI traceable thermal diffusivity value of IG-110 as a reference material.

Thermal diffusivity on the laser flash method is determined from a specimen thickness and a heat diffusion time. And thermal diffusivity values of materials are a function of temperature. On the other hand, the measurement system is also composed of three units corresponding to each quantities, length, time, and temperature. Then we checked and calibrate our measurement system and estimated the uncertainty of a measurement result. In this paper, evaluation of uncertainty of thermal diffusivity measurements in the case of IG-110 specimen is reported.

2. EXPERIMENTAL

2.1. Specimens

IG-110 is a grade of isotropic high-density graphite manufactured by Toyo Tanso Co., Ltd., and was selected as a candidate for a thermal diffusivity reference material. About 100 rods of IG-110, which are 100 mm in length and 10 mm in diameter, are stocked in NMIJ. We sampled a rod from our stocks. The bulk density of this rod is 1.76 $Mg\cdot m^{-3}$ and the electrical resistance is 1050 μ Ω according to the manufacturer.

We prepared a set of specimens, which consist of four specimens with 10 mm in diameter and 1.4, 2.0, 2.8 and 4.0 mm in thickness cut out from adjacent position of one rod in order to determine the thermal diffusivity value independent of the specimen thickness. These specimens are polished to make both surface parallel. The thickness variation of a specimen is several micrometers. These processes are necessary to define the specimen thickness with a small uncertainty.

2.2. Measurements

The thickness of the specimens is measured using a linear gauge. For a specimen, we measure five points as shown in Fig.1. We determine the specimen thickness as the average of the values of five points.

Thermal diffusivity measurement carried out using a laser flash measurement system. The block diagram is shown in Figure 2. This measurement system includes some technical improvements in order to make thermal diffusivity measurements under well-defined initial and boundary conditions as follows:

(i) uniform pulse heating of a specimen by an improved laser beam using an optical fiber (reduction of the nonuniform heating error) [2,5];

(ii) development of a fast infrared radiation thermometer with an absolute temperature scale (reduction of the nonlinear temperature detection error) [2,6]; and

(iii) introduction of a new data analysis algorithm, "a curve-fitting method", where the entire regions of the temperature history curve is fitted by a theoretical solution under the real boundary condition (reduction of the heat loss error) [2,7].

A curve-fitting method [2,7] is used to determine the thermal diffusivity from the temperature history curve obtained by the laser flash measurement, as shown in Fig. 3. The entire set of experimental data is fitted by Cape and Lehman's theoretical curve [8] corrected by Josell et al. [9], which gives an analytical solution under the heat loss boundary condition. Both the thermal diffusivity and the Biot number are simultaneously determined by this curve fitting method. The origin of the time was set at the center of gravity of the observed laser-pulse intensity distribution when the observed temperature history curve is fitted to a theoretical curve [10].

Thermal diffusivity values were measured with changing heating laser pulse energies at a constant effective specimen temperature. A unique thermal diffusivity value can be determined for homogeneous materials independent of measurement conditions by extrapolating to zero heating laser pulse energy on the plot of apparent thermal diffusivity values measured with the laser flash method as a function of heating laser pulse energy [3]. Figure 4 exhibits heating laser pulse energy dependence of thermal diffusivity at room temperature for an IG-110 specimen set. Horizontal axis represents amplitude of output signal of infrared radiation thermometer. Lines are the best fit to all data points. The intrinsic thermal diffusivity is the value extrapolated to zero amplitude of the output signal. This figure shows that the thermal diffusivity values of the different thickness specimens from the same rod agree within about 5 %. Measurements were carried out from room temperature to about 1200 K for these four specimens. The temperature dependence of thermal diffusivity of this IG-110 specimen set is plotted as shown in Fig. 5. Deviation of thermal diffusivity values between four specimens is enough small at high temperature.

3. EVLUATION OF UNCERTAINTY

Laser flash measurement is observation of one-dimensional thermal diffusive phenomena. Because of this simply phenomena and popularity, laser Flash Method is known as a reliable method. However, it is difficult to make an ideal condition of theoretical model at an actual measurement. For example, there is a heat loss effect and a non-uniform effect. It is important that we acknowledge some problems and check repeatability and accuracy of measurement.

According to the half time method [1],

$$\alpha(T) = 0.1388 \times \frac{d^2}{t_{1/2}},$$

where, a is the thermal diffusivity of the specimen which is a property dependent on temperature. T is the temperature of the specimen, d is the specimen thickness, and $t_{1/2}$ is the half time. This equation means that thermal diffusivity is determined from length, time and temperature. As a measurement, we measure a specimen thickness using a gauge and exactly estimate a heat diffusion time using a laser flash instrument. And a temperature is determined by temperature sensor such as a thermocouple. In fact, the measurement system is composed of three units corresponding to each quantities, length, time, and temperature as shown in Fig.2. According to this, we check a measurement system and estimate an uncertainty.

3.1. Calibration and examination of a measurement system

The measurement system consists of three units corresponding to length, time and temperature, as shown in Fig. 2. We checked them and calibrated traceably to the national standard.

The linear gauge for measuring specimen thickness is calibrated using gauge blocks with 1.0mm, 2.0mm and 4mm in thickness.

Sampling frequency of the data acquisition was checked by the function generator, which was calibrated by the frequency measurement division in NMIJ. The time lag between the data acquisition part and the function generator was estimated about 0.0001 %.

The temperature measurement part was also calibrated. The working standard thermocouple was calibrated at four melting fixed points (In, Al, Zn, Cu). We calibrated the reference thermocouple compared with the working standard thermocouple. The temperature scale of the laser flash measurement system was corrected considering temperature gradient around the sample holder using the reference thermocouple. All thermocouples are R type.

3.2. Uncertainty

Major sources of uncertainty in thermal diffusivity measurements are as follows: (i) uncertainty of specimen thickness, (ii) uncertainty of time scale, (iii) uncertainty of infrared radiation thermometry [6], (iv) uncertainty of pulse width [10,11], (v) non-uniform heating effect [2,12,13], (vi) heat loss effect [7,8,9], (vii) drift of the specimen temperature, (viii) uncertainty of data analysis, (ix) uncertainty of extrapolating analysis, and (x) uncertainty of the specimen temperature measurement. Considering these sources and following "Guide to the expression of uncertainty in measurement" (GUM) [14], we have made a preliminary evaluation of the uncertainty of the measurement.

3.2.1 Uncertainty of specimen thickness

Specimen thickness was measured at room temperature using a linear gauge calibrated by block gauges. Combined standard uncertainty of specimen thickness is calculated from uncertainty of length of block gauge, linear gauge calibration and deviation of five measured-value of specimen thickness in a specimen. Since the thermal diffusivity value is proportional to square of the specimen thickness, the relative uncertainty of thermal diffusivity attributed to the specimen thickness is 2 times of the combined standard uncertainty of the specimen thickness.

3.2.2 Uncertainty of sampling time

Thermal diffusivity is calculated from heat diffusion time determined from a measured temperature history curve. It is important that measurements are carried out on condition that the time-frequency resolution enough high to analysis the temperature history curve. The temperature history curve is recorded during from twice of the half time before pulse heating to 18 times of the half time after pulse heating in our measurement as shown in Fig.3. Uncertainty of heat diffusion time is attributed to arbitrary of analysis of temperature history curve and uncertainty of time scale of the measurement. The former is discussed in 3.2.8. The latter is checked using a function generator traceable to the national standard according to 3.1.

The overall accuracy of the time interval of the data acquisition is estimated from uncertainty of frequency of the function generator and frequency deviation between the function generator and the data acquisition part. This is about 0.0001 %. Additionally uncertainty of finite data sampling is the ratio of the sampling time of A/D conversion to the half time.

3.2.3 Uncertainty of infrared radiation thermometry

NMIJ have developed the infrared radiation thermometer for laser flash method [2,5]. A response time of this infrared radiation thermometer is as fast as 10 microseconds. Uncertainty of thermal diffusivity caused by response time of the infrared radiation thermometry is evaluated as the ratio of the response time to the half time.

3.2.4 Uncertainty of pulse width

In the theoretical model, a heating pulse shapes the delta function. It is necessary to correct pulse width in the actual measurement with a finite pulse width. The origin of the time was set at the center of gravity of the observed laser-pulse intensity distribution when the observed temperature history curve is fitted to a theoretical curve [7,10]. The accuracy of the origin of time for data analysis attributed to pulse width contributes to uncertainty of thermal diffusivity.

3.2.5 Non-uniform heating effect

The spatial energy distribution of the pulsed laser beam is observed using a beam profile instrument. The laser beam was uniform enough that uncertainty caused by the non-uniform heating effect is about 1 % [13].

3.2.6 Heat loss effect

Experimental curves were fitted by the theoretical function proposed by Cape and Lehman and corrected by Josell *et al*. [8, 9]. The function considers a heat loss effect. It is known that the uncertainty of approximation of this theoretical function is about 5 % of heat loss effect [7]. We have considered that uncertainty due to heat loss effect is 5 % of deviation between thermal diffusivity value estimated from the half time method without heat loss [1] and that from the curve fitting method [7].

3.2.7 Distortion of a temperature history curve by drift of specimen temperature

Generally, a measurement starts at a condition when the specimen temperature is almost stable. However, there is possibility that the specimen temperature distorts a little during a measurement with one pulse heating. A little distortion becomes an uncertainty factor. Then we assumed that the drift of specimen temperature equals to 0.01 K during the measurement over 20 times of the half time. In the case that the specimen temperature at the end of a measurement linearly increase (or decrease) 0.01 K rather than that at the start of a measurement, uncertainty due to distortion of a temperature history curve by drift of specimen thickness estimated about 0.03 % from an analysis of a temperature history curve with totally 0.01 K sloped background with about 4 K amplitude of output signal.

3.2.8 Uncertainty of data analysis

We analyse measurement data using the curve fitting method as shown in Fig. 3 [7]. There are two fitting parts for the curve fitting analysis, a temperature increase part and a temperature decay part. How choose them becomes uncertainty factor. Uncertainty of data analysis estimated as variation of results with shifted fit part.

A temperature history curve is data acquired from twice of a half time before pulse heating to 18 times of a half time after pulse heating in our measurement. Empirically, we choose fitting area as follows;

(i) A fitting part in a temperature increase part with about 0.4-1.0 times of a half time in width selected in the range between 0.5 and 2 times of a half time around the half time.

(ii) A fitting part in a temperature decay part with about 2-4 times of a half time in width selected in the range between 6 and 12 times of a half time.

3.2.9 Uncertainty of extrapolating analysis

An intrinsic thermal diffusivity is determined as an intercept of extrapolating to zero energy [3]. We usually measure more than 20 times for a specimen at a constant temperature changing pulsed laser energy. Then we fitted a linear function to a series of measurement includes these about 20 data. The standard deviation of data from the linear function is also obtained. This is uncertainty of extrapolating analysis.

3.2.10 Uncertainty of specimen temperature

A specimen temperature is detected using a thermocouple installed in the specimen holder. The temperature scale of the measurement system with the specimen holder is calibrated according to 3.1. Uncertainty of specimen temperature measurement is combined uncertainty of the working standard thermocouple, the reference thermocouple and the comparative calibration of the temperature scale of the system. It, on the other hand, takes about two hours to obtain enough data determining one intrinsic thermal diffusivity value similarly to 3.2.9. The specimen temperature fluctuate about

0.4 K during the measurements at room temperature. Uncertainty of specimen temperature is evaluated from these two kinds of uncertainties.

Then we can estimate uncertainty of thermal diffusivity due to specimen temperature uncertainty in the case of a specimen of which temperature dependence is known. Now we discuss the case of an IG-110 isotropic graphite. The Debye temperature of Graphite is known about 2000 K. Exponential decay increasing temperature of thermal diffusivity dominant in this temperature range. We have assumed an exponential function and obtain the temperature dependence for this IG-110 specimen set as shown in Fig. 4 and 5. According to the temperature dependence function, magnitude of the thermal diffusivity change due to uncertainty of specimen temperature is estimated. Uncertainty of thermal diffusivity arise from specimen temperature uncertainty is provided from that magnitude and a standard deviation of the exponential function.

Finally, combined standard uncertainty of thermal diffusivity measurement is the square root of the sum of these 10 factors. The expanded uncertainty of the thermal diffusivity measurement at room temperature is estimated to be about 4 % with the coverage factor k = 2. Table 1 shows an example of an error budget table on laser flash thermal diffusivity measurement for an IG-110 specimen at room temperature [15].

4. CONCLUSION

We have been studying the laser flash method in order to establish an SI traceable thermal diffusivity standard. This time we carried out an uncertainty evaluation on the laser flash measurement in order to determine the thermal diffusivity value of IG-110 as a candidate reference material.

Thermal diffusivity values of materials are function of temperature. Thermal diffusivity is calculated from the specimen thickness and the heat diffusion time at a fixed temperature observed by the laser flash method. Since the measurement system is composed of three units corresponding to length, time and temperature, we checked and calibrated our measurement system traceably to the national standard of each quantity and estimated the uncertainty of a measurement result.

In the case of an IG-110 specimen set, the relative expanded uncertainty of thermal diffusivity with the coverage factor $k = 2$ is about 3 - 6 % over the temperature range from room temperature to about 1200 K.

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Figure Captions

Fig. 1. (a) A specimens set of IG-110 graphite. These are cut from near place in a same rod. (b) the 5 points for the specimen thickness measurement.

Fig. 2. Schematic diagram of measurement system. This measurement system is composed of three units corresponding to each quantities, length, time, and temperature. We checked and calibrated them, respectively.

Fig. 3. The procedure of the curve-fitting method to analyse a temperature history curve observed the laser flash measurement.

Fig. 4. Heating laser pulse energy dependence of thermal diffusivity at various temperatures for the IG-110 specimen set at room temperature. Horizontal axis represents amplitude of output signal of infrared radiation thermometer. Dashed lines are the best fit to all data points. An intrinsic thermal diffusivity is determined by extrapolating to zero amplitude of the output signal along these lines. This figure indicates that the intrinsic thermal diffusivity can be estimated at each temperature.

Fig. 5. Temperature dependence of thermal diffusivity on an IG-110 specimens set with 1.4, 2.0, 2.8, 4.0 mm in thickness. Dashed lines are the best fit to all data points. The temperature dependence dominates a function of exponential decay.

Table Caption

Table. 1. An example of error budget table on the laser flash thermal diffusivity measurement at room temperature for an IG-110 specimen.

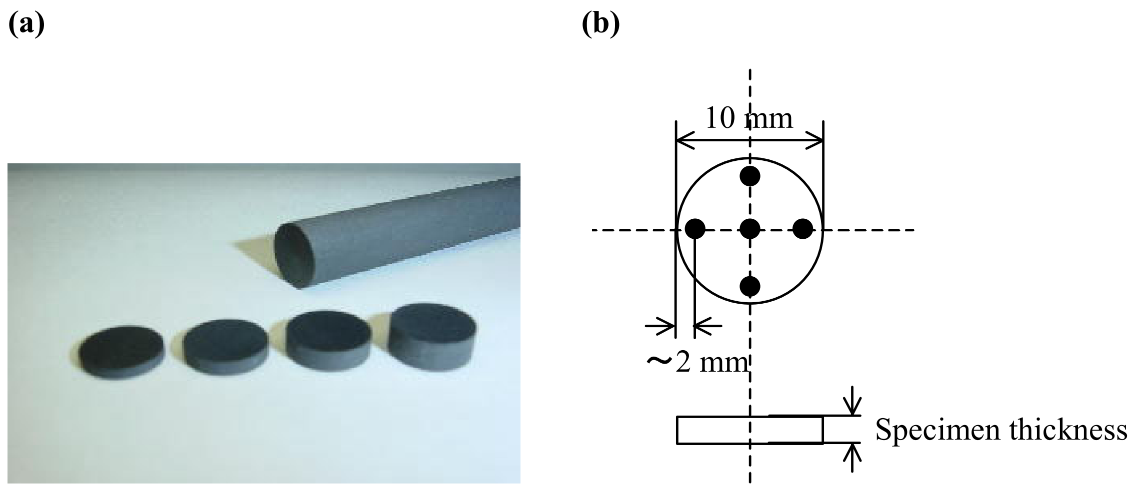


Figure .1

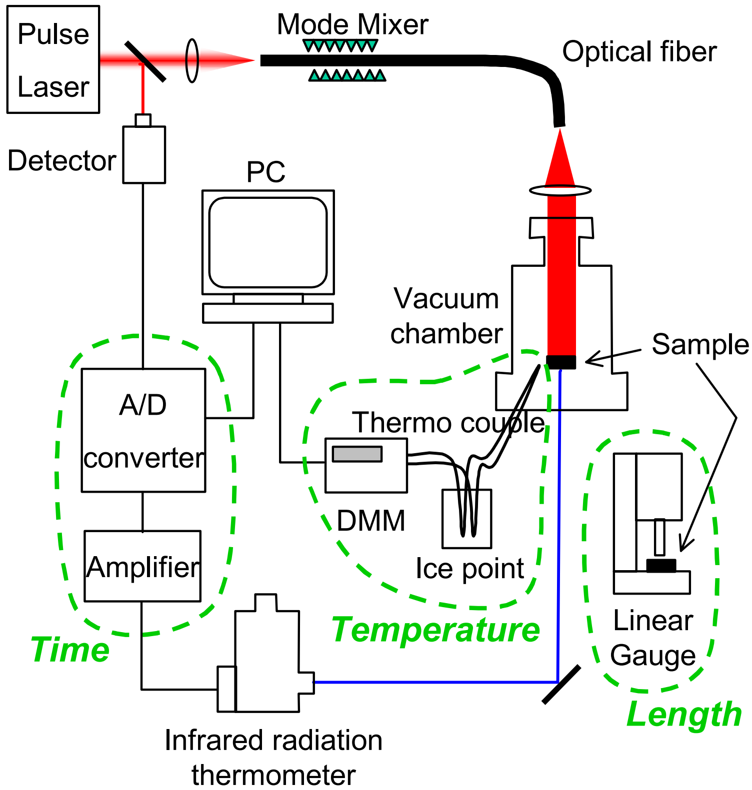


Figure. 2

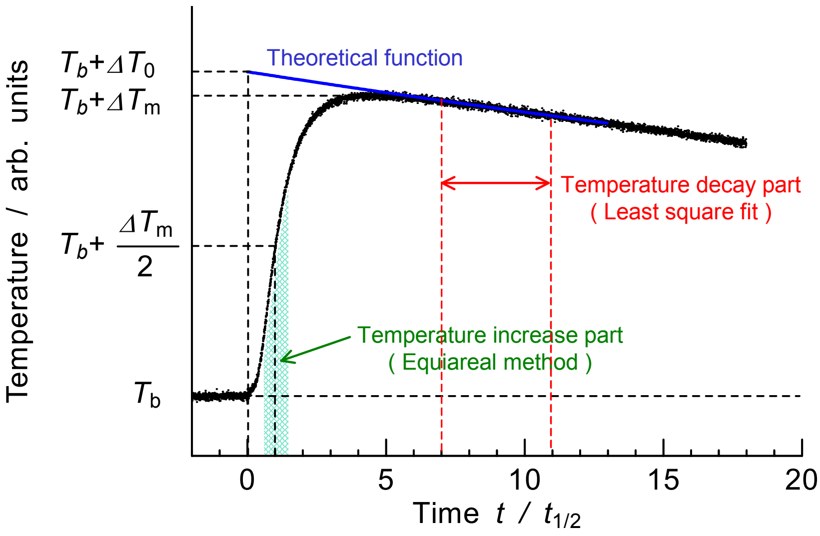


Figure. 3

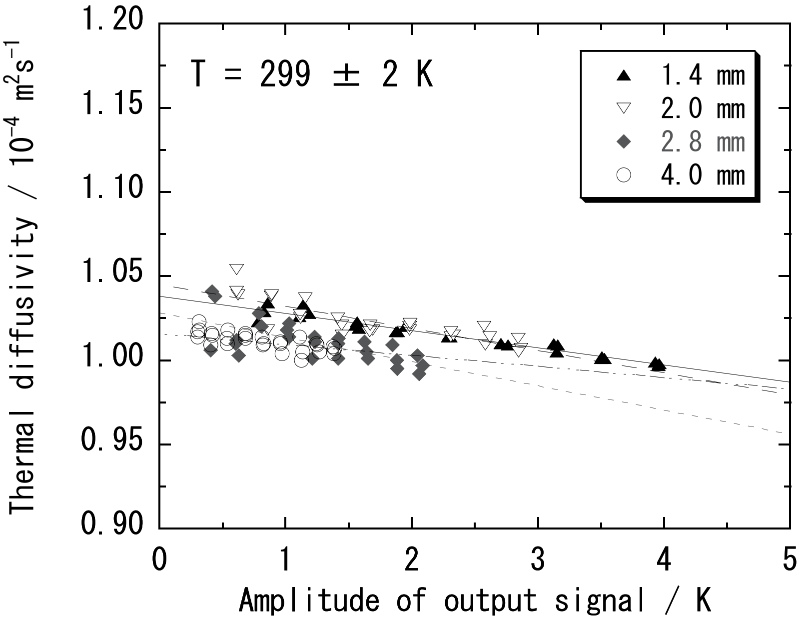
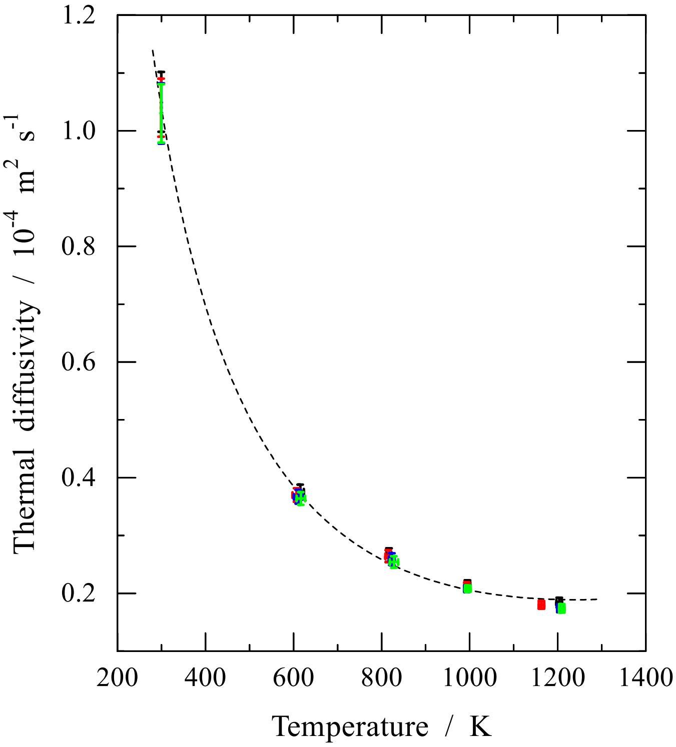


Figure. 4



1.2
1.0
0.8
0.6
0.4
0.2
Thermal diffusivity / 10^{-4} m^2 s^{-1}
200
400
600
800
1000
1200
1400
Temperature / K

Figure. 5

Table. 1

Uncertainty of thermal diffusivity measurement					
Factor of uncertainty	Type	Value of uncertainty	Standard uncertainty	Relative uncertainty %	Combined relative uncertainty %
Specimen thickness Block Gauge: $u(L_{BG})$ Calibration of a linear gauge: $u(L)$ Standard deviation of an average on measured specimen thickness: $u(d_M)$	 B B A	 1.2×10^{-8} m 9.5×10^{-8} m 7.0×10^{-6} m	7.0×10^{-6} m	0.4	
Sampling time Frequency of a function generator : δf_i Phase shift of recorded signal : Δf_M A/D conversion: $t_s / N_{1/2}$	 B B B	 1.3×10^{-8} % 1.3×10^{-4} % 1.6×10^{-6} %	1.2×10^{-4} %	0.0001	
Infrared radiation thermometry Temporal response time for $t_{1/2}$: $t_{IR} / t_{1/2}$	B	1.0×10^{-5} s	2.1×10^{-4} %	0.0001	
Pulse width Deviation depends on origin time: $\Delta\alpha_{t0}(u(t_0)) / \alpha_{1/2}$	B	1.2 %	0.6 %	0.7	1.7
Non-uniform heating effect Non-uniform heating effect : $\Delta\alpha_{NU}/\alpha_{1/2}$	B	1.8 %	1.0 %	1.0	
Heat loss effect Heat loss effect: $\delta\alpha_{hl}/\alpha_{CF}$	B	0.8 %	0.4 %	0.5	
Distortion of a temperature history curve drift of specimen temperature : $\delta\alpha_{dr}/\alpha_m$	B	0.2%	0.1 %	0.1	
Analysis of temperature history curve Selection of fitting parameters : $\delta\alpha_0$	A	0.4 %	0.2 %	0.2	
Extrapolating analysis Standard deviation of a function: $\delta(SD_{ex})$	A	1.6 %	0.2 %	0.9	
Uncertainty of effective specimen temperature measurement					
Factor of uncertainty	Type	Value of uncertainty	Standard uncertainty	Combined standard uncertainty	Combined relative uncertainty %
Temperature scale of a calibrated thermocouple Uncertainty value from certification sheet: u_{10}	B	0.4 K	0.4 K		
Temperature scale of thermocouple Comparative calibration of thermocouple1: u_{TC1} Comparative calibration of thermocouple2: u_{TC2}	 A A	 0.9 K 1.3 K	1.6 K	1.7 K	
Stability of an effective specimen temperature Fluctuation of effective specimen temperature: $\delta(SD_{Tb})$	A	0.4 K	0.4 K		1.0
Temperature dependence of thermal diffusivity Standard deviation of a function: δf	A	1.0 %	1.0 %	1.0 %	
Combined standard uncertainty ($k = 1$)					2.1
Relative expanded uncertainty ($k = 2$)					4.1