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Study on a Thermal-diffusivity Standard for Laser Flash Method Measurements[1]

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The National Metrology Institute of Japan (NMIJ) of AIST has been studying the laser flash method in order to establish an SI traceable thermal-diffusivity standard. Key technologies have been developed to reduce the uncertainty in laser flash measurements. In the present study, an uncertainty evaluation has been carried out on the laser flash measurement method in order to determine the thermal diffusivity value of IG-110, a grade of isotropic high-density graphite, as a candidate reference material. The thermal diffusivity measured by the laser flash method is derived from a specimen thickness and a heat diffusion time. And a laser flash measurement is carried out at a given temperature. The measurement system is composed of three sections corresponding to each measured quantity: length, time, and temperature. Therefore, we checked and calibrated our measurement system, and estimated the uncertainty of measurement results for the case of a grade of isotropic graphite.

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

1. INTRODUCTION

The flash method is one of the most popular methods to measure the thermal diffusivity of solid materials above room temperature [1]. The National Metrology Institute of Japan (NMIJ) of AIST has been studying the laser flash method in order to establish a reference material as a

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thermal-diffusivity standard [2, 3]. A reference material is useful to verify the reliability of laser flash instruments. To reduce the uncertainty in laser flash measurements [2], we have developed key technologies, including uniform pulse heating on specimens, development of a fast infrared radiation thermometer, introduction of a new data analysis algorithm, and an extrapolation method to determine an intrinsic thermal-diffusivity value. We have also investigated candidate reference materials for laser flash measurements based on the following concepts:

(1) It has good homogeneity and stability.
(2) It can be measured without black coatings.
(3) There is a set of specimens of different thicknesses from the same substance.
(4) The uncertainty of the thermal diffusivity value is evaluated.
(5) The thermal diffusivity value of a set is SI traceable.

From our research, it has been found that IG-110 is appropriate as a reference material for laser flash measurements. IG-110, a grade of isotropic graphite manufactured by Toyo Tanso Co., Ltd, is black. It has good homogeneity and stability [3, 4]. From measurements with changing pulse-heating energies, it is confirmed that the thermal-diffusivity values of IG-110 specimens with different thicknesses 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.

The thermal diffusivity using the laser flash method is determined from a specimen thickness and a heat diffusion time. The thermal diffusivity of materials is a function of temperature. The measurement system is composed of three sections corresponding to measured quantities of length, time, and temperature. Therefore, we checked and calibrated our measurement system and estimated the uncertainty of a measurement result. In this paper, an evaluation of the uncertainty of thermal-diffusivity measurements, for the case of IG-110 specimens, 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 at NMIJ.

We sampled a rod from our stocks. The bulk density of this rod is $1.76\,\mathrm{Mg\cdot m^{-3}}$, and the electrical resistance is $1050\mu\Omega$ according to the manufacturer.

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

2.2. Measurements

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

Thermal diffusivity measurements were carried out using the laser flash measurement system as shown in Fig. 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 non-uniform heating error) [2, 5]; (ii) development of a fast infrared radiation thermometer with an absolute temperature scale (reduction of the non-linear temperature detection error) [2, 6]; and (iii) introduction of a new data analysis algorithm, "a curve-fitting method," where entire regions of the temperature history

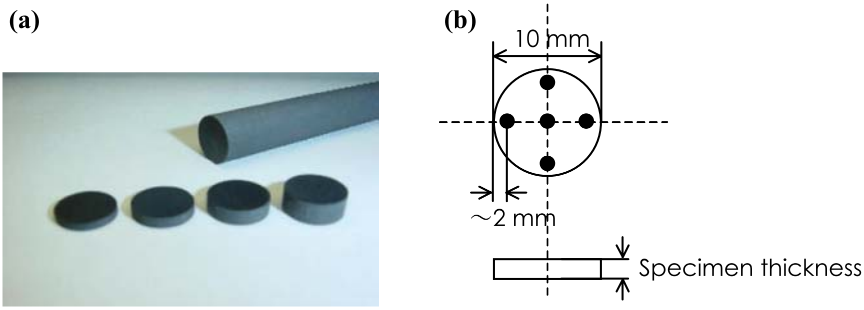

Fig. 1. (a) Specimen set of IG-110 graphite (these are cut from near the same place in a given rod) and (b) five points for the specimen thickness measurements.

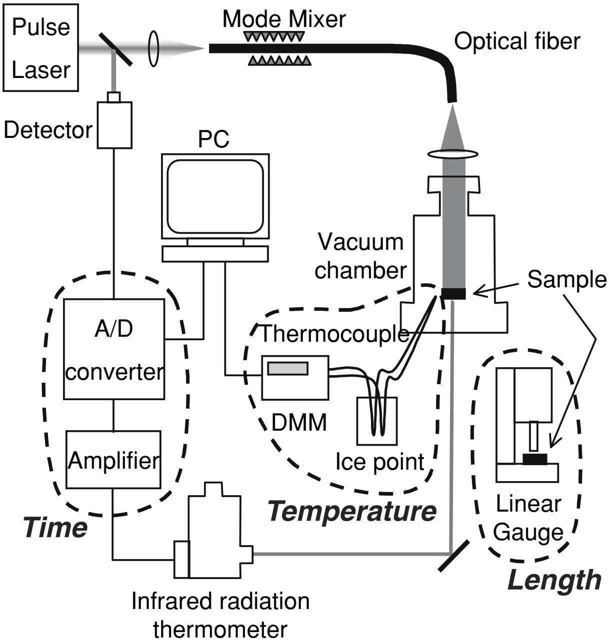

Fig. 2. Schematic diagram of the measurement system. This measurement system is composed of three sections corresponding to the measured quantities of length, time, and temperature.

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

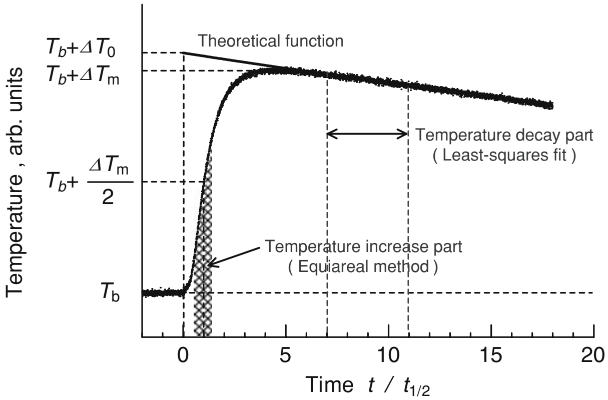

Fig. 3. Procedure of the curve-fitting method to analyze a temperature history curve observed by the laser flash measurement.

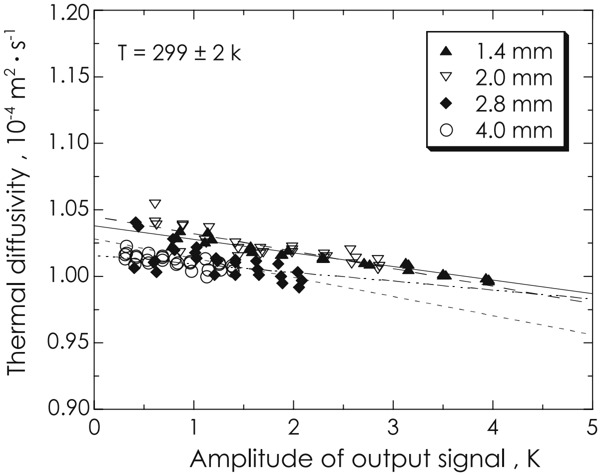

Fig. 4. Heating laser pulse energy dependence of thermal diffusivity values at various temperatures for the IG-110 specimen set at room temperature. Horizontal axis represents the amplitude of the output signal of the infrared radiation thermometer. Dashed lines are the best fit to all data points. An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

[3]. Figure 4 exhibits the heating laser pulse energy dependence of the thermal diffusivity at room temperature for an IG-110 specimen set. The horizontal axis represents the amplitude of the output signal of an infrared radiation thermometer. The lines are the best fit to all data points for respective specimens. The intrinsic thermal diffusivity is the value

extrapolated to the 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 thermal diffusivity of each specimen at each temperature above room temperature was obtained using the extrapolation analysis in the same way as Fig. 4. The temperature dependence of the thermal diffusivity of this IG-110 specimen set is plotted as shown in Fig. 5. Figure 5 shows that the values of the thermal diffusivity agree within about 7% at above room temperature.

In the present study, we calculated thermal-diffusivity values with the specimen thicknesses measured at room temperature. The laser flash measurement determines the heat diffusion time. The thermal diffusivity value is calculated from the specimen thickness and the heat diffusion time. If thermal expansion data of the material are available, we can cor-

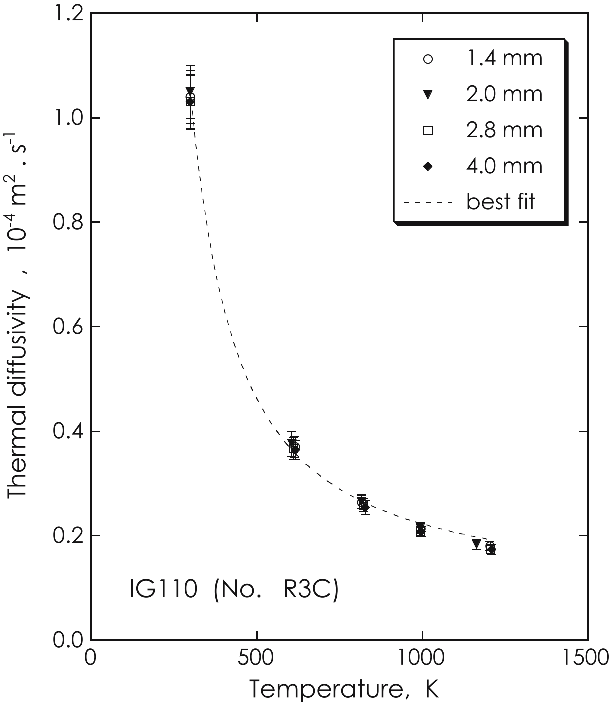

Fig. 5. Temperature dependence of thermal diffusivity values for an IG-110 specimen set with thicknesses of 1.4, 2.0, 2.8, and 4.0 mm. Dashed line is the best fit of all data points with the exponential function in Section 3.2.10.

rect the specimen thickness and re-calculate the absolute value of thermal diffusivity if necessary.

3. EVALUATION OF UNCERTAINTY

The laser flash measurement method involves observations of one-dimensional thermal diffusive phenomena. Because of this simple experimental design and its popularity, the laser flash method is considered reliable. However, it is difficult to satisfy ideal conditions of the theoretical model with the actual measurements. For example, there are a heat loss effect and a non-uniform heating effect. It is important to check the repeatability and accuracy of the measurements.

The half-rise time method [1] to derive the thermal diffusivity is expressed as

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

where α is the thermal diffusivity of the specimen which is dependent on temperature, T is the temperature of the specimen, d is the specimen thickness, and $t_{1/2}$ is the half-rise time defined by the interval required for the back-surface temperature to reach one-half of the maximum temperature rise. This equation means that the thermal diffusivity is determined from measurements of length, time, and temperature. For a measurement, we measure a specimen thickness using a gauge and estimate a heat diffusion time using a laser flash instrument. The temperature is determined using a temperature sensor such as a thermocouple. In fact, the measurement system is composed of three sections corresponding to the parameters of length, time, and temperature as shown in Fig. 2. Accordingly, we tested the measurement system and estimated an uncertainty.

3.1. Calibration and Examination of the Measurement System

The measurement system consists of three sections corresponding to length, time, and temperature, as shown in Fig. 2. We checked them and calibrated the system with traceability to national standards. A linear gauge for measuring the specimen thickness was calibrated using gauge blocks with thicknesses of 1.0, 2.0, and 4.0 mm.

The sampling frequency of the data acquisition module was checked using a function generator, which was calibrated by the Frequency Measurement Division of NMIJ. The time lag between the data acquisition part and the function generator was estimated to be about 0.0001%.

The temperature measurement section consists of the working thermocouple, a digital multimeter and an automatic ice point as shown in Fig. 2. The working thermocouple is Type-R (Pt/Pt13%Rh). The junction of the working thermocouple is located within about 3 mm of the specimen.

The temperature measurement section was calibrated as follows: we set a 4.0 mm thickness specimen with a 2.0 mm hole at the center and inserted the junction of the reference thermocouple into the hole. The reference thermocouple is also Type-R (Pt/Pt13%Rh). The specimen and the reference thermocouple were put in the specimen holder in our measurement system. The working thermocouple was compared with the reference thermocouple set with the specimen. Calibration was carried out under this configuration from room temperature to about 1200 K. The temperature difference between the specimen and the junction of the working thermocouple is compensated by this calibration procedure. The reference thermocouple is calibrated by comparison with the standard thermocouple using a thermocouple calibration furnace in our laboratory. The standard thermocouple is also Type-R (Pt/Pt13%Rh) enclosed in a high purity alumina protection tube of 6 mm diameter that was calibrated at 4 fixed melting points (In, Al, Zn, Cu) by an accredited calibration laboratory of the Japan Calibration Service System (JCSS).

3.2. Uncertainty

The major sources of uncertainty in thermal-diffusivity measurements are as follows: (i) uncertainty of specimen thickness, (ii) uncertainty of time scale, (iii) uncertainty of response time of infrared radiation thermometry [6], (iv) uncertainty of pulse width [10, 11], (v) non-uniform heating effect [2, 12, 13], (vi) heat loss effect [7–9], (vii) drift of the specimen temperature, (viii) uncertainty of data analysis, (ix) uncertainty of extrapolation analysis, and (x) uncertainty of the specimen temperature measurement. Considering these sources and following the "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

The specimen thickness was measured at room temperature using a linear gauge calibrated by block gauges. The combined standard uncertainty of the specimen thickness is calculated from the uncertainty of the length of a block gauge, the linear gauge calibration, and the deviation of five measured values of the specimen thickness. Since the thermal-diffusivity value is proportional to the square of the specimen thickness, the relative uncertainty of the thermal diffusivity attributed to the specimen thickness is a factor of two larger than the combined standard uncertainty of the specimen thickness.

3.2.2. Uncertainty of Sampling Time

The thermal diffusivity is calculated from the heat diffusion time determined from a measured temperature history curve. It is important that measurements are carried out at the condition where the time-frequency resolution is sufficiently high to analyze the temperature history curve. The temperature history curve is recorded during the period from twice the half-rise time before pulse heating to 18 times the half-rise time after pulse heating is initiated in our measurement as shown in Fig. 3. The uncertainty of the heat diffusion time is attributed to an analysis of the temperature history curve and the uncertainty of the time scale of the measurement. The former is discussed in Section 3.2.8, and the latter was checked using a function generator traceable to national standards discussed in Section 3.1.

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

3.2.3. Uncertainty of Response Time of Infrared Radiation Thermometry

NMIJ has developed an infrared radiation thermometer for the laser flash method [2, 5]. The response time of this infrared radiation thermometer is as fast as 10μs. The uncertainty of the thermal diffusivity resulting from the response time of the infrared radiation thermometer is evaluated as the ratio of the response time to the half-rise time.

3.2.4. Uncertainty of Pulse Width

Ideally, a surface of a specimen is instantaneously heated by the delta function pulsed laser at the measurement starting time $t = 0$. However, the laser pulses in the actual measurements have finite pulse width. It is proposed that a real pulse with finite width can be approximated by a delta function at $t = 0$ as the real pulse width is relatively short compared to the half-rise time [10].

In our measurements of IG-110, the pulse width is about 300μs and the half-rise time is longer than about 2 ms. Since the half-rise time is longer than five times the pulse width, the starting 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 starting time for data analysis attributed to the pulse width contributes to the uncertainty of the 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 sufficiently uniform that the uncertainty caused by the non-uniform heating effect is about 1% [2, 5, 13].

3.2.6. Heat Loss Effect

Experimental curves were fitted by the theoretical function proposed by Cape and Lehman [8] and corrected by Josell et al. [9]. The function takes the heat loss effect into consideration. It is known that the uncertainty of approximation of this theoretical function is about 5% of the heat loss effect [7]. We have considered that the uncertainty due to the heat loss effect is 5% of the deviation between the thermal diffusivity values estimated from the half-rise time method without heat loss [1] and those 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 a possibility that the specimen temperature drifts slightly during a measurement after one pulse heating. The shift may distort the temperature. We estimated that the drift of the specimen temperature is less than 0.01 K during the measurement over a period of 20 times the half-rise time. For the case when the specimen temperature at the end of a measurement increases (or decreases) linearly by 0.01 K rather than at the start of a measurement, the uncertainty due to the distortion of a temperature history curve by a drift of the specimen temperature was estimated to be about 0.03% from an analysis of the temperature history curve with a total drift temperature of 0.01 K for an output signal about 4 K amplitude.

3.2.8. Uncertainty of Data Analysis

We analyzed the data using the curve fitting method as shown in Fig. 3 [7]. There are two parts for the curve-fitting analysis: a temperature increasing part and a temperature decay part. The uncertainty of the data analysis is estimated as a variation of the results with a shift of fitting parts. The data for temperature history curve is acquired from twice the half-rise time before pulse heating to 18 times the half-rise time after pulse heating in our measurements. Empirically, we choose the fitting area as follows;

(i) a temperature increasing part with about 0.4–1.0 times the half-rise time in width was selected in the range between 0.5 and 2 times the half-rise time about the half-rise time.

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

3.2.9. Uncertainty of Extrapolation Analysis

An intrinsic thermal diffusivity is determined as an intercept by extrapolating to zero energy [3]. It is also effective to reduce the deviation of thermal diffusivity due to non-linearity of spectral radiance on temperature associated with the radiation thermometry. We usually carry out measurements more than 20 times for a specimen at a constant temperature with changing pulsed laser energies. Then we fitted a linear function to these data points. The standard deviation of data points from the linear function is obtained. This is taken as the uncertainty of the extrapolation 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 as mentioned in Section 3.1. The uncertainty of the specimen temperature measurement is the combined uncertainties of the working standard thermocouple, the reference thermocouple, and the comparative calibration of the temperature scale of the system. On the other hand, it takes about 2 h to obtain enough data for determining one intrinsic thermal diffusivity value as mentioned in Section 3.2.9. The specimen temperature fluctuates approximately 0.4 K during the measurements at room temperature. The uncertainty of the specimen temperature is evaluated from these two kinds of uncertainties.

We can estimate the uncertainty of the thermal diffusivity due to the specimen temperature uncertainty for the case of a specimen for which the temperature dependence is known. Now we discuss the case of IG-110 isotropic graphite. The Debye temperature of graphite is known to be about 2000 K [15]. The thermal diffusivity decreases exponentially in this temperature range as the temperature increases. The transport of heat by phonons is dominant in this temperature region. We have assumed an exponential function,

$$\alpha(T) = M_1 + M_2 \exp\left(\frac{M_3}{T}\right),$$

where α is the thermal diffusivity, T is the temperature, and $M_n (n = 1, 2, 3)$ is constant. And we have obtained the temperature dependence for this IG-110 specimen set as shown in Figs. 4 and 5. According to the temperature dependence function, the magnitude of the thermal-diffusivity change due to the uncertainty of the specimen temperature is estimated. The uncertainty of thermal diffusivity arising from the specimen temperature uncertainty is determined by the magnitude and standard deviation of the exponential function.

Finally, the combined standard uncertainty of thermal-diffusivity measurements is the square root of the sum of the squares of uncertainties due to these 10 factors discussed above. The expanded uncertainty of the thermal-diffusivity measurement at room temperature is estimated to be about 4% with a coverage factor $k = 2$. Table I shows an example of an uncertainty budget for laser flash thermal-diffusivity measurements for an IG-110 specimen at room temperature [16].

4. CONCLUSION

We have studied the laser flash method in order to establish an SI traceable thermal diffusivity standard. In the present study, we carried out an uncertainty evaluation on the laser flash measurement in order to determine the thermal diffusivity of IG-110 as a candidate reference material.

Thermal diffusivity values of materials are a function of temperature. The thermal diffusivity is calculated from the specimen thickness and the heat diffusion time at a fixed temperature as determined with the laser flash method. Since the measurement system is composed of three sections corresponding to length, time, and temperature measurements, we checked and calibrated our measurement system with traceability to national standards for each quantity and estimated the uncertainty of a measurement result.

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

Table I. Example of Uncertainty Budget for Laser Flash Thermal-diffusivity Measurements at Room Temperature for an IG-110 Specimen

Uncertainty of thermal diffusivity measurement					
Factor of uncertainty	Type	Value of uncertainty	Standard uncertainty	Relative uncertainty (%)	Combined relative uncertainty (%)
Specimen thickness					
Block Gauge	B	1.2×10^{-8} m			
Calibration of a linear gauge	B	9.5×10^{-8} m	7.0×10^{-6} m	0.4	
Standard deviation of an average on measured specimen thickness	A	7.0×10^{-6} m			
Sampling time					
Frequency of a function generator	B	1.3×10^{-8}%			
Phase shift of recorded signal	B	1.3×10^{-4}%	1.2×10^{-4}%	0.0001	
A/D conversion	B	1.6×10^{-6}%			
Response Time of Infrared radiation thermometry					
Temporal response time for $t_{1/2}$	B	1.0×10^{-5} s	2.1×10^{-4}%	0.0001	1.7
Pulse width					
Deviation depends on origin time	B	1.2%	0.6%	0.7	
Non-uniform heating effect					
Non-uniform heating effect	B	1.8%	1.0%	1.0	
Heat loss effect					
Heat loss effect	B	0.8%	0.4%	0.5	
Distortion of a temperature history curve					
Drift of specimen temperature	B	0.2%	0.1%	0.1	
Analysis of temperature history curve					
Selection of fitting parameters	A	0.4%	0.2%	0.2	
Extrapolation analysis					
Standard deviation of a function	A	1.6%	0.2%	0.9	

Table I. (*Continued*)

Uncertainty of effective specimen temperature measurement					
Factor of uncertainty	Type	Value of unvertainty	Standard uncertainty	Combinded standard Uncertainty	Combined relative uncertainty (%)
Temperature scale of a working standard thermocouple					
Uncertainty value stated in certification sheet	B	0.4 K	0.4 K		
Temperature scale of thermocouple					
Comparative calibration of reference thermocouple	A	0.9 K	1.6 K		
Comparative calibration of the system	A	1.3 K		1.7 K	1.0
Stability of an effective specimen temperature					
Fluctuation of effective specimen temperature	A	0.4 K	0.4 K		
Temperature dependence of thermal diffusivity					
Standard deviation of a function	A	1.0%	1.0%	1.0%	
Combined standard uncertainty ($k = 1$)					2.1
Relative expanded uncertainty ($k = 2$)					4.1

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REFERENCES

1. W. J. Parker, R. J. Jenkins, C. P. Butler, and G. L. Abbott, *J. Appl. Phys.* **32**:1679 (1961).
2. T. Baba and A. Ono, *Meas. Sci. Technol.* **12**:2046 (2001).
3. M. Akoshima and T. Baba, *Int. J. Thermophys.* **26**:151 (2005).
4. M. Akoshima and T. Baba, *Proc. 28th ITCC* (2005).
5. T. Baba, M. Kobayashi, A. Ono, J. H. Hong, and M. M. Suliyanti, *Thermochim. Acta* **218**:329 (1993).
6. M. Kobayashi, T. Baba, and A. Ono, *Jpn. J. Thermophys. Prop.* **8**:143 (1994).
7. A. Cezairliyan, T. Baba, and R. Taylor, *Int. J. Thermophys.* **15**:317 (1994).
8. J. A. Cape and G. W. Lehman, *J. Appl. Phys.* **34**:1909 (1963).
9. D. Josell, J. Warren, and A. Cezairliyan, *J. Appl. Phys.* **78**:6867 (1995).
10. T. Azumi and Y. Takahashi, *Rev. Sci. Instrum.* **52**:1411 (1981).
11. R. E. Taylor and J. A. Cape, *J. Appl. Lett.* **5**:212 (1964).
12. J. A. McKay and J. T. Schriempf, *J. Appl. Phys.* **47**:1668 (1976).
13. T. Baba, *Proc. 17th Jpn. Symp. Thermophys. Props., Tsukuba* (1996), p. 379.
14. BIPM, IEC, IFCC, ISO, IUPAP, and OIML, *Guide to the Expression of Uncertainty in Measurement* (ISO, Geneva, Switzerland, 1995).
15. B. T. Kelly, *Physics of Graphite* (Applied Science Pubs., London, 1981), p. 153.
16. M. Akoshima, T. Baba and H. Kato, *Proc. 24th Jpn. Symp. Thermophys. Props., Nagano* (2004), p. 261.