Title: Metrological Evaluation of a High Temperature Laser Flash Thermal Diffusivity Instrument

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Abstract

This work describes a general metrological characterization of a high temperature – high pressure thermal diffusivity measurement instrument, using NIST SRM 8425 sample. Sources of measurement errors were analyzed and the results were subjected to rigorous statistical evaluation, to determine the uncertainty associated with the thermal diffusivity measurements for the instrument used. Results consisting of 264 data points covering the temperature range from 20 ^{o}C to 1000 ^{o}C are presented. The analysis showed that the relative standard deviation of the mean thermal diffusivity values varied from 0.1 % to 0.5 %. The combined standard uncertainty associated with the measurements was determined to be 1.127 %, and the expanded uncertainty $\pm$ 2.25 %, for 95 % confidence level.

Key Words: thermal diffusivity, errors, uncertainty

Introduction

The flash method for measuring thermal diffusivity has been used increasingly ever since its introduction in the late 50's, by Parker et al [1]. Today, this is the method of choice to determine not only thermal diffusivity, but also thermal conductivity and specific heat capacity for temperature regions where steady state methods no longer function, and for new or exotic materials, where available geometry precludes conventional direct methods. The theory, operating principles, and analytical data evaluation have been amply described in the literature [1-10]. Excellent metrological evaluations have been presented by Suliyanti et al [11] and Cezairliyan et al [12]. This work complements and further reinforces the prior data.

Method and Equipment

The flash method is based on applying a very short energy pulse on the front face of a small disc shaped specimen, and calculating its thermal diffusivity from the characteristic curve (thermogram) of the temperature excursion of its rear face. Parker derived [1] that the specimen's thermal diffusivity, α, can be calculated for ideal conditions from:

$$\alpha = 0.1388 \frac{L^2}{t_{1/2}} \tag{1}$$

where L is its thickness and $t_{1/2}$ is a characteristic time on the thermogram, when the rear surface reaches one half of its final temperature. Numerous corrections have been introduced to account for radiative heat losses during the process, the finite width of the laser pulse, the non-uniform heating of the sample, and other factors interfering with the experiment, to more realistically represent actual experimental conditions [2-10].

The test equipment today invariably consists of a laser or other source of a short high energy pulse, a furnace to contain the specimen, and an infrared detection system with high speed recording capabilities, to record the rear face temperature rise. The equipment used for this study, a high temperature – high pressure system, able to perform measurements from 20 ºC to 1000 ºC, and from vacuum to 30 bar internal pressure, has been previously described [13]. The performance of the instrument was analyzed as part of a large testing program performed on a graphite reference material (NIST SRM 8425). The results are presented below together with a thorough analysis of the sources of errors present in the measurement process.

General Considerations on Measurement Errors and Standard Uncertainty

The difference between the measured value of a property and its true value is called measurement error. Any measurement is affected by errors, due to a multitude of factors: the imperfection of the instruments used and of the measurement methods followed, the influence of external perturbations, the variation of the ambient conditions, the subjectivity of the operator, etc. In certain cases, applying an adequate correction can eliminate the errors.

On the other hand, the true value associated with the property to be measured, as well as the measurement error, are idealized notions and can not be determined. It is only the result of the measurement that becomes known, but the result itself is worthless if no information is available on how correctly it describes that property. For this purpose, the *measurement uncertainty* is the most widely used and generally accepted concetp, essentially different from the concept of *error*. A somewhat “traditional” definition of the uncertainty of measurement is the following: “an estimate characterizing the range of values within which the true value of a measurand lies” [VIM – International Vocabulary of Basic and General Terms in Metrology, 1st edition, ISO, Geneva, 1984]. A newer version eliminates any reference to the “true value” (which is an unknowable quantity): “a parameter associated with the result of the measurement, that characterizes the dispersion of the values that could reasonably be attributed to the measured property” [VIM, 2nd edition, 1993; 17].

The uncertainty has different components, which may be categorized according to the method used to evaluate them. Each component of the uncertainty, however evaluated, is represented by an estimated standard deviation called *standard uncertainty*. When a statistical analysis of a series of observations is possible, the method of evaluation is called “of type A”. When means other than statistical analysis are used, the evaluation is referred to as “of type B”.

The type A component of the uncertainty, also known as (experimental) standard deviation of the mean, is determined based on the results obtained by repeating the same measurement, in the same conditions. Obviously, such an evaluation can not render evident uncertainty components arising from systematic effects. Therefore, in most cases, the type B evaluation is unavoidable, consisting in the identification of the individual sources of measurement errors, evaluation of their contribution to the measurement process in the form of “uncertainty components” and combining them to obtain the “type B component” of the uncertainty. Data from previous measurements, technical specifications of the instruments used, general knowledge related to the property measured, uncertainties reported in certificates used, can be taken into consideration. In general terms, the uncertainty components are either obtained from an outside source (books, certificates, manufacturer's specifications, etc.), or from an assumed distribution (normal, rectangular, triangular, etc.) In this latter case, the quantity in question is modeled by a probability distribution, and lower and upper limits are estimated, such that the best estimated value of the input quantity lies between these limits, with a particular probability, which is called *confidence level*.

It is important to underline that there is no difference in the nature of the uncertainty components resulting from the two types of evaluation, both of them being quantified by standard deviations. Therefore, the "type A" and the "type B" components can be combined, generating the *combined standard uncertainty* of the measurement, to provide an interval of measurement results that is expected to encompass a large fraction of the possible values, by multiplying the combined standard uncertainty with a *coverage factor* (usually in the range 2 to 3) [16-18], to obtain the value of the *expanded uncertainty*. Either the confidence level or the value of the coverage factor chosen has to be specified in addition to the expanded uncertainty range.

The expanded uncertainty represents the preferred parameter to characterize the uncertainty of measurements in most industrial and commercial applications, with the value $k = 2$ for the coverage factor, equivalent to a 95 % confidence level when a normal distribution of the measurement results may be assumed. This the approach that will be followed in the present paper, according to the principles of uncertainty evaluation and expression outlined in [16-18].

Metrological Characterization of Equipment

To determine the performance characteristics of the equipment repeated tests had to be performed on a stable and well-characterized specimen. The graphite reference material NIST SRM 8425 was chosen for this task. Twenty four tests were performed on a specimen in nitrogen atmosphere, from 20 ºC to 1000 ºC, every 100 ºC at 1 bar pressure.

SRM 8425 is a fine grain, nearly isotropic graphite, with density and electrical resistivity variation related to position within the mother block, a shortcoming that limits its usefulness in absolute terms. The thermal diffusivity values considered “certified” for this material derive from the results of a large international round-robin testing program which took place over a period of 19 years and whose results were published in 1984 [14], considered accurate within ± 10 %.

The results of the tests performed, as well as the certified values of thermal diffusivity for this material, are summarized in table I and figure 1.

The data reported in the Average column contain the mean values of the twenty four tests performed at each temperature. Each individual thermal diffusivity value was calculated as the average of three values, derived using the Clark and Taylor correction applied for three different ratios of partial times (required for reaching the particular percentages of the temperature increase on the thermogram): 80/20, 70/30 and 67/33.

Table I Thermal diffusivity values for SRM 8425

TEMPERATURE (ºC)	THERMAL DIFFUSIVITY Average of Measured Values (cm^2/s)	Standard Deviation (Absolute) (cm^2/s)	Standard Deviation (Relative) (%)	NIST Data (cm^2/s)	Percent Dev. from NIST Values (%)
30	0.7872	0.00360	0.457		
100	0.6260	0.00143	0.228	0.5990	4.51
200	0.4713	0.00036	0.076	0.4575	3.02
300	0.3708	0.00037	0.099	0.3662	1.26
400	0.3056	0.00030	0.099	0.3040	0.53
500	0.2619	0.00043	0.166	0.2596	0.89
600	0.2291	0.00067	0.292	0.2267	1.06
700	0.2049	0.00057	0.278	0.2015	1.69
800	0.1854	0.00266	0.144	0.1818	1.98
900	0.1704	0.00017	0.100	0.1661	2.56
1000	0.1579	0.00033	0.209	0.1534	2.89

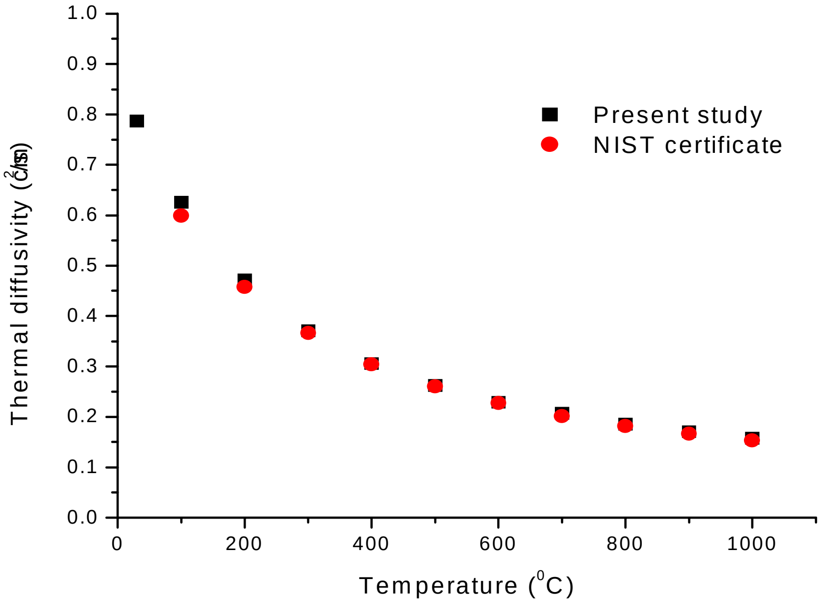

Figure 1 *Thermal diffusivity values for SRM 8425 (average of 24 tests), compared with NIST certified values.*

Estimate of Uncertainty

In principle, the thermal diffusivity is obtained from the thickness of the specimen and from a characteristic time function describing the propagation of heat from the front surface of the specimen to its back surface. The sources of uncertainties in the measurement are associated with the sample itself, the temperature measurements, the performance of the detector and the data acquisition board, the data analysis and more specifically the finite pulse time effect, the nonuniform heating of the sample, and radiative heat losses. These sources can be considered of systematic nature, and will be discussed individually. The random component of the uncertainty will be derived separately, from the repeated measurements performed on the graphite reference.

Type A Component Uncertainty

The relative standard deviation values in table I represent the type A uncertainties involved in this measurement process. The highest value (0.46 %) corresponds to room temperature determinations, all others being smaller than 0.3 %. This was expected, since low temperature determinations involve considerable smaller “half-max time” values than those at higher temperatures, which leads to an increase in the uncertainty associated with them. It can be concluded that 0.5 % is well representing the type A component of the uncertainty associated with the thermal diffusivity measurements.

Type B Component of Uncertainty

Type B components of uncertainty will be discussed individually for each source of such uncertainties. Estimates of these components will be given as limits between which the particular influence quantity may generate a variation of the measured value. This means that a rectangular distribution is implicitly assumed for the occurrence probability of the values within the limits given.

Specimen Thickness

Thermal diffusivity is related to the square of the specimen thickness, thus $\Delta\alpha/\alpha=2\Delta l/l$. For the typical 3.5 mm thick specimen, for example, if the room temperature thickness is measured with ± 4 μm, the error limits associated with the thickness measurement are ± 0.2 %.

The material's thermal expansion during the test introduces another source of error, which could be corrected if an additional measurement of thermal expansion for the entire temperature range of interest could be performed. No such test was performed on the NIST reference. However, thermal expansion values for generic AXM 5Q type graphite were found [15] and an average coefficient of linear thermal expansion of $2 \cdot 10^{-6}$ / ºC was representative for the temperature range 20 ºC … 1000 ºC. Therefore, the error implied by not correcting the results for thermal expansion is estimated to be ± 0.2 %.

Specimen Temperature

The steady state temperature of the specimen does not enter into the equation generating the thermal diffusivity values. A particular criteria in the instrument's software is set to define the thermal equilibrium conditions necessary before starting the test. Constant sample temperature within 1 ºC for minimum 5 minutes was found to be an acceptable condition. The temperature was measured using a type K thermocouple (nickel – chromium / nickel – aluminum), whose accuracy is specified by its manufacturer as ± 1.1 ºC or ± 0.4 % of the measured value. It should be noted that during the thermal diffusivity experiment, the sample's effective temperature is higher than its steady state temperature, due to the pulse generated by the laser. The uncertainty in thermal diffusivity resulting from not knowing the true sample temperature is estimated to be less than ± 0.3 %.

Performance of the Detector

The manufacturer of the detector specifies the maximum frequency of the signal that can be detected as 5 MHz, which is equivalent with 0.07 μs response time. When testing a graphite specimen, the usual "half-max time" values are approximate 20 ms for room temperature measurements and 1 s for 1000 ºC. In each of these extreme cases, the uncertainty in thermal diffusivity due to the response time of the detector is extremely small, actually considerably smaller than 0.01 %. In practice, this source of error is considered negligible.

The signal response of the detector is linear to the input radiation for small temperature changes (of the order of 10 ºC). An important factor in the measurement is to ensure that the detector is operating within its linear range. The nonlinearity is attributed mainly to the nonlinear dependence of the Planck's formula on temperature. The maximum transient temperature rise at the back surface of the sample is usually less than 10 ºC for temperatures up to 1000 ºC. Planck's formula can be approximated by a linear function, with a discrepancy less than 1 %, within this small temperature change. The solution for avoiding using the detector outside its linear range is to use the smallest laser power that generates a measurable temperature rise to the back face of the sample. All the measurements performed in this work involved the minimum possible power level for the laser, generating less than 3 ºC temperature rises. The error limits of the thermal diffusivity due to the nonlinearity of the detector are estimated to be less than ± 0.5 %.

Performance of the Digital Data Acquisition Board

The operational characteristics of the instrument’s data acquisition board are as follows:

Signal resolution: 16 bit (1 in 65,535, or 0.0015 %)
Maximum sweep length: 30,000 points in 2 s, followed by 7,500 points in 10 s
Time base accuracy: 0.01 % (specified by the manufacturer)
Overall accuracy: 0.01 % (specified by the manufacturer)
Linearity: 0.005 % (specified by the manufacturer)

Based on these characteristics, it is considered that the errors in thermal diffusivity due to the digital data acquisition board’s operational errors are negligible small compared to the other error sources.

Uncertainty of the Time Origin

Since the measurement of thermal diffusivity is, in essence, a time measurement, it is important to know as good as possible where the time origin lies. For the instrument used for this work, the laser pulse width, measured at half of its maximum value, is 250 μs. Azumi and Takahashi [7] recommend approximating the pulse by a delta function and considering its center of gravity as being the origin of time for the experiment. For measurements performed on graphite specimens, for example, the “half max time” values measured are approximate 20 ms for 20 ºC, and close to 1 s for 1000 ºC. Because the laser pulse is very narrow, not correcting the data for the pulse width does not have a large influence. The worst possible case is for measurements performed at low temperatures, where the error limits in thermal diffusivity associated with the time origin are estimated to be ± 0.6 %.

Nonuniform Heating Effect

The uniformity of the laser beam is directly related to the uniformity of the sample's heating, and is usually a major source of error in the measurements. One of the great advantages of the equipment used for this work, in comparison with others, is the use of optical fiber for delivering the laser pulse to the sample's chamber. This leads to a very high uniformity of the laser beam, practically eliminating any problems associated with nonuniform heating. The error limits in thermal diffusivity due to this effect are estimated to be less than ± 0.1 %.

Radiative Heat Loss

During a laser flash experiment, radiative heat losses from the specimen are unavoidable. The specimen may lose heat by radiation from its front, back and side surfaces. The losses are very small at low temperatures, but can increase considerably with the temperature. The contribution of the radiative heat losses is expressed by nondimensional parameters called Biot numbers, Y, defined as $Y = 4 \cdot \varepsilon_s \cdot \sigma T_0^{3} \cdot r / \lambda$, with $0<Y<1$, where ε_s is the hemispherical emissivity of the specimen, σ is the Stefan-Boltzmann constant, T_0 is the steady state temperature of the specimen, r is its radius, and λ is the thermal conductivity. For the sides of the sample, the losses are much smaller than those from the two faces, due to the very small gap existing between the specimen and its holder.

A computer simulation program was used to estimate the influence of the radiative heat losses on the thermal diffusivity. Assuming that the experiment is using a 3.5 mm thick specimen, and maintaining all the test parameters the same, different Biot numbers were assigned to each face of the specimen (front, back and side). The configurations used were:

- all three Biot numbers equal to 0.1;
- the front face Biot number 0.2, the back and the side one 0.1;
- the front and back face Biot numbers 0.2, and the side one 0.1;
- the front face Biot number 0.3, the back face 0.2, and the side one 0.1;
- the front face Biot number 0.4, the back face 0.2, and the side one 0.1.

The simulation was run for samples with thermal diffusivity values from 0.5 to 0.2 cm^2/s. In all cases, the variation in thermal diffusivity due to the changes in the heat losses (monitored through the Biot numbers), was within $\underline{+}$ 1.5 %, decreasing with the thermal diffusivity. It was concluded that the error limits in thermal diffusivity due to the radiative heat losses may be estimated as 1.5 %.

The various uncertainties estimated are summarized in table II, along with the correspondent standard deviations. Since a rectangular distribution was assumed for the occurrence probability of the values within the given limits, the individual standard deviations, u_i, can be calculated as:

$$u_i = \frac{a}{\sqrt{3}} \qquad (2)$$

where $\underline{+}\ a$ are the limits of the estimated uncertainty for each case.

Table II Sources of type B uncertainties

Source of Uncertainty	Estimated Uncertainty Limits (%)	Standard Deviation (%)
Specimen's thickness	± 0.2	0.115
Specimen's thermal expansion	± 0.2	0.115
Specimen's temperature	± 0.3	0.173
Detector's performance	± 0.5	0.289
Data acquisition board's performance	0.0	0.000
Laser pulse width	± 0.6	0.346
Nonuniform specimen heating	± 0.1	0.058
Radiative heat losses	± 1.5	0.866

Since no significant correlation between the input quantities involved in the various sources of type B uncertainties may be assumed, the type B component of the uncertainty in the thermal diffusivity measurement can be calculated as the square root of the sum of the squares of the above tabulated standard deviation values. This leads to 1.01 % type B uncertainty associated with the experiments. The combined standard uncertainty is calculated similarly from the type A and B components. With 0.5 % type A uncertainty, the combined standard uncertainty becomes 1.127 %. The expanded uncertainty can be calculated by multiplying the combined standard uncertainty values with the coverage factor $k = 2$, which corresponds to a confidence level of 95 %. For the present evaluation, this leads to an expanded uncertainty of ± 2.25 % within 95 % confidence level.

This is in excellent agreement with the results published by Sulyianti, Baba and Ono [11] listing 2.3 to 2.7 % uncertainty using pyroceram 9606, a material that is lower in diffusivity and therefore slightly more prone to heat loss errors. Another similar study, by Cezairliyan, Baba and Taylor [12] concluded a 3 % uncertainty for POCO AXM 5Q1 material (generically identical to the material used in this work). No expanded uncertainty was given in either case, and no confidence levels were defined.

Conclusions

This study presents a metrological characterization of a high temperature – high pressure thermal diffusivity equipment, through a series of tests performed on a NIST SRM 8425 specimen. The results of a large testing program are presented, along with a detailed analysis of the sources of errors present and the correspondent uncertainties.

It is concluded that the expanded uncertainty associated with the thermal diffusivity results of measurements performed on the FlashLine™ 5000 instrument [19] used for this study, is ± 2.25 % within 95 % confidence level.

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