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JRP 17IND11 Hi-TRACE

Industrial process optimisation through improved metrology of thermophysical properties

Good practice guide on thermal diffusivity measurements by the laser flash method up to 3000 °C

Lead Participant: LNE (B. Hay)

Other Participants: ArianeGroup, CEA, FHWS, Netzsch, NPL, VINS

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1. Scope

Measurement practices in the laboratories for determining the thermal diffusivity of solid materials are in many cases based on national or international standards. The most important standards related to this topic are given below:

- *EN 821-2 (1997) "Advanced technical ceramics - Monolithic ceramics - Thermo-physical properties - Part 2: Determination of thermal diffusivity by the laser flash (or heat pulse) method"*
- *EN 1159-2 (2004) "Advanced technical ceramics - Ceramic composites - Thermophysical properties - Part 2: Determination of thermal diffusivity"*
- *ISO 18755 (2005) "Fine ceramics (advanced ceramics, advanced technical ceramics) - Determination of thermal diffusivity of monolithic ceramics by laser flash method"*
- *ASTM E1461-13 (2013) "Standard Test Method for Thermal Diffusivity by the Flash Method"*
- *ISO 22007-4 (2017) "Plastics - Determination of thermal conductivity and thermal diffusivity - Part 4: Laser flash method"*
- *ISO 19629 (2018) "Fine ceramics (advanced ceramics, advanced technical ceramics) - Thermophysical properties of ceramic composites - Determination of unidimensional thermal diffusivity by flash method"*

These standards describe the general methodology to be applied to measure the thermal diffusivity according to the laser flash method, without giving specific recommendations for measurements performed at very high temperature. The purpose of this guide is to outline the basic technical requirements for performing thermal diffusivity measurements of solid materials at high temperature (between 1500 °C and 3000 °C) by applying the laser flash method. The aim of this document is not to replace or harmonize the existing standards, but it could serve as a reference for future standards revisions.

2. General comments

The laser-flash method is by far the most frequently used method to measure the thermal diffusivity of solid materials [1]. In this method, a thin disk specimen is maintained at a uniform temperature in a furnace operating either by resistive or inductive heating. One side of the specimen is then subjected to a short thermal excitation generated by a laser pulse, which has a uniform spatial energy distribution. The induced transient temperature rise on the opposite side is measured versus time with an infrared detector. Examples of laser flash experiment are presented in the figure 1 for resistive or inductive heating.

The thermal diffusivity is obtained by comparing the measured temperature-time curve (thermogram) with a theoretical model describing the transient heat conduction through the specimen. Various models and identification methods can be used to calculate the thermal diffusivity from the measured thermograms. The original method proposed by Parker [1], which assumes ideal conditions (adiabatic experiment, energy uniformity and infinitesimal duration of the laser pulse, homogenous and opaque specimen), shall not be applied in particular at high temperature. Alternative methods enabling correction for non-ideal and boundary conditions are more appropriate, in particular at high temperatures, and shall be used (more details can be found in the CEN and ISO standards listed in section 1).

The following sections give specific recommendations and advices for thermal diffusivity measurements with the laser flash method above 1500 °C to 3000 °C.

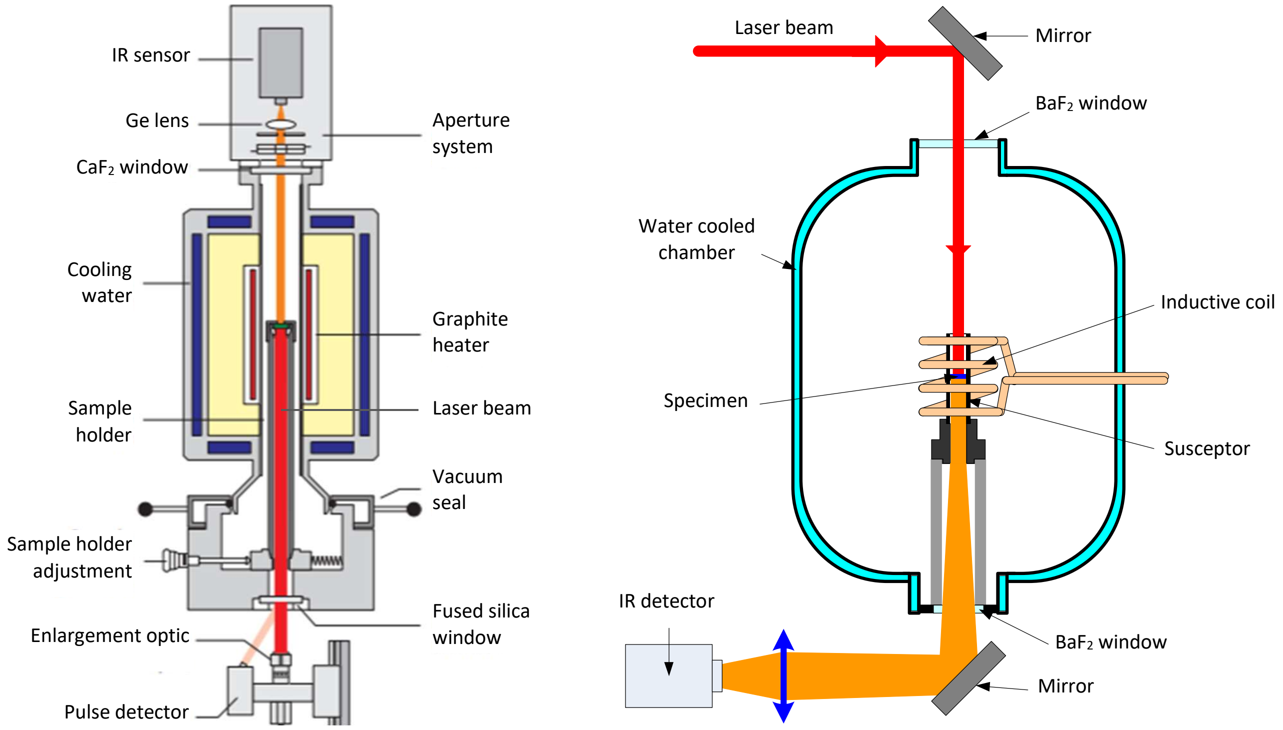

Figure 1 Schematic diagram of a laser flash experiment with resistive (left) or inductive (right) heating

3. Requirements for the Laser Flash Apparatus

3.1 Furnace

The test chamber shall be either a resistive or an inductive furnace capable of operation within the required temperature range. It shall be equipped with two windows, one transparent to the pulse radiation and the other transparent to the working wavelength range of the IR detector measuring the thermogram. BaF_2 windows are recommended for thermal diffusivity measurements performed at very high temperature, as their transmission is higher than 90 % from 0.25 µm to 10 µm (CaF_2 windows with transmission higher than 90 % from 0.4 µm to 8 µm can also be used as alternative).

Tests can be performed either under vacuum or under inert gas. Even if measurements under vacuum will reduce convection losses, it is recommended to work with inert gas at very high temperature (argon or helium up to 2000 °C, and preferably helium above 2000 °C) to avoid sublimation phenomena of the tested specimens. In that case, the specimen shall be in a horizontal position in order to reduce convection effects of the gas on it.

Additional precautions shall be taken if an inductive system is used to heat the specimen. The specimen shall be put in a graphite holder (also named susceptor) which is heated by induction thanks to a coil connected to a high frequency generator. A filter shall be put at the output of the high frequency generator in order to reduce the high frequency electromagnetic interferences on the signal delivered by the IR detector. The inductive coil, which is usually made of copper, shall be coated with an electrically insulating ceramic deposit in order to avoid the appearance of sparks between the susceptor and the coil that can damage the susceptor and disturb the measurements.

3.2 Specimen holder

The specimen holder shall hold the specimen stable with minimum thermal contact to limit conductive heat losses, and shall be designed to suppress stray light from the laser beam being transmitted to the IR detector. An example of specimen holder is shown in figure 2.

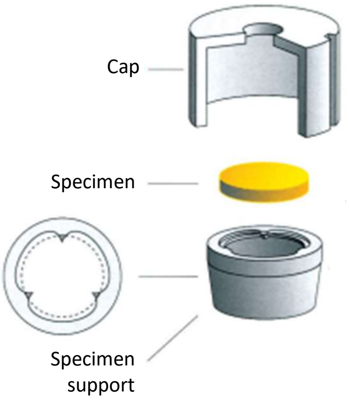

Figure 2 Example of specimen holder

Depending on the tested material, the specimen holder should be made of either compatible refractory metallic materials (if the tested material is a pure metal or a metallic alloy) or graphite to limit chemical interactions between the holder and the specimen at high temperatures. To avoid contact between a metallic specimen and a graphite sample holder, an alternative solution is to put a thin washer made of molybdenum or tungsten between the specimen and the holder. If available, it is recommended to consult relevant material phase diagrams when choosing the appropriate type of material for the specimen holder.

3.3 Detector

The transient temperature rise of the specimen rear side shall be measured optically with an infrared detector, which shall be carefully chosen depending on the temperature range. In contrast to the usually applied HgCdTe IR detectors for lower temperatures, in particular InGaAs and Si detectors are recommended for test temperatures ranging from 1500 °C and 3000 °C.

3.4 Radiation pyrometer

The steady-state temperature of the specimen shall be measured before the laser pulse with infrared radiation thermometers (or pyrometers) having an operating temperature range covering that of the furnace. Bi-chromatic radiation thermometers shall be used preferably as in that case the knowledge of the emissivity of the specimen is not needed. However even the application of bi-chromatic radiation thermometers need careful evaluation, because rapid changes in emissivity might critically affect the measured temperature [2].

Even if the sensitivity of the thermal diffusivity to temperature is low at high temperature, the error on the temperature measurements by radiation thermometry in laser flash apparatus that can be higher than 50 °C at 3000 °C if the pyrometers are not calibrated, has an influence on the uncertainty of measurement. These radiation thermometers, which can be subjected to drift with time, need to be periodically calibrated to ensure trueness of temperature measurements.

This temperature calibration can be done in-situ by using metal-carbon eutectic high temperature fixed points (HTFP) having known freezing temperatures. The cross-section of a small HTFP crucible (main part of the cell and cap) is shown in figure 3 (right). In the temperature calibration configuration, the laser is disabled and the radiation thermometer aims at the aperture of the HTFP blackbody cavity positioned in the furnace at the location of the specimens (cf. figure 3 - left). The HTFP cell is thermally stabilized at a temperature higher than the freezing point of the metal carbon eutectic used. The temperature of the furnace is then rapidly lowered to a temperature below the freezing point by reducing the power delivered in the inductive coil or in the resistance, causing thus the cooling of the HTFP. The calibration of the radiation pyrometer is performed by comparing the value of the freezing temperature measured and the reference value. More details about this procedure can be found in [3].

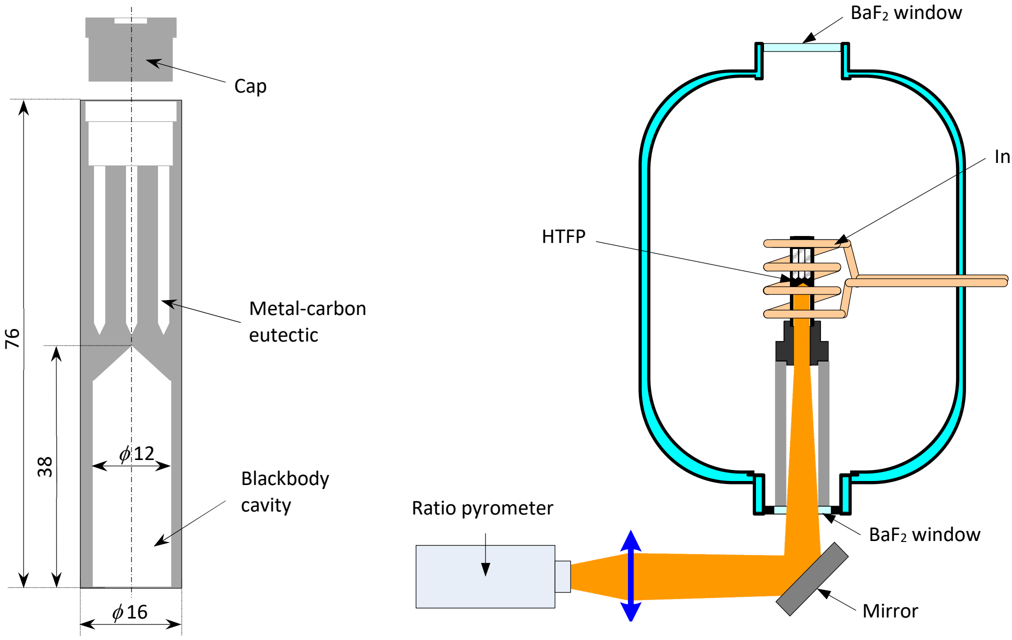

Figure 3 In-situ calibration of radiation thermometer using small HTFP

4. Preparation of the test

4.1 Test specimen preparation

The specimen is a disk whose faces perpendicular to the measurement shall be flat and parallel. The parallelism error of the two faces shall be less than 0.05 mm. The specimen thickness shall be larger than 1 mm (thickness of 2 mm or 3 mm seems to be a good compromise), because deformation of the specimens can be observed at high temperature if the specimen is too thin.

The specimen shall be representative of the material being examined and shall be prepared and handled with care (preferably with gloves) in order to avoid any contamination.

It can be sometimes needed to sandblast, grind or graphite coat the specimen surface in order to increase its emissivity and therefore to improve the signal-to-noise ratio of the infrared detector output. The use of graphite coating can have however influence on the measurements due to potential interaction with the specimen at high temperature.

4.2 Cleaning of the furnace

It is recommended to clean the furnace and the sample holder after a series of measurements above 2000 °C. Particles can indeed be released from the pieces of the furnace and the sample holder at these high temperatures when they are made of graphite, and can then settle on the windows or contaminate the tested specimens.

In case of inductive heating, the presence of graphite particles in the furnace drastically increase the risk of sparks between the susceptor and the coil at very high temperature.

5. Procedure of measurement

As for the previous sections, the general recommendations given in the CEN and ISO standards listed in section 1 regarding the measurement procedures of thermal diffusivity shall be applied. The present section gives additional advices for thermal diffusivity measurements performed from 1500 °C to 3000 °C.

5.1 Measurements

- Measure the thickness and the mass of the specimen before starting the test.
- It is recommended to perform three consecutive measurements at each level of temperature under repeatability conditions, the average of the three obtained results giving the thermal diffusivity value.
- The measurements shall be performed at 23 °C and at different levels of temperature covering the investigated temperature range when heating-up and/or cooling-down the specimen. This series of measurement shall be repeated at least during a second heating/cooling cycle and a final measurement shall be done at 23 °C.
- Measure the thickness and the mass of the specimen after each thermal cycle.

The aim of this procedure is to check the stability of the material when heated at high temperature, and to detect potential modifications of the specimen microstructure (which can be different depending on the heating/cooling rates and the holding times at high temperatures).

5.2 Data analysis

The calculation of the thermal diffusivity value from the obtained experimental thermogram is performed according to one of the methods described in the CEN and ISO standards listed in section 1. This thermal diffusivity value $a_0(T)$ is calculated using the thickness of the specimen d_0 measured before the test at room temperature with a calibrated micrometre.

The correct thermal diffusivity value $a(T)$ based on the specimen thickness d_T at the measurement temperature T is determined with the following equation.

$$a(T) = \left(\frac{d_T}{d_0}\right)^2 \cdot a_0(T)$$

The higher the temperature, the more important is this correction. It has been shown in [4] that the error on thermal diffusivity values is about 3 % for measurements performed on molybdenum and tungsten respectively at 2200 °C and 2400 °C and about 4 % for measurements carried out at 3000 °C on graphite specimens, if the thermal expansion is neglected.

6. Uncertainty of measurement

The relative expanded uncertainty associated with thermal diffusivity measurement shall be calculated in accordance with the ISO/BIPM Guide to the expression of uncertainty in measurement [5] by considering the following sources of uncertainties:

- Measurement of the physical quantities involved in the determination of thermal diffusivity (thickness and temperature of the specimen, output voltage of the infrared detector, time base).

  Uncertainties associated with the measurements of these physical quantities result mainly from the combination of the uncertainties due to the noise of measurement, the resolution, and calibration of the equipment. Another source of uncertainty could be a possible drift of the baseline of the signal delivered by the IR detector due to electromagnetic disturbances induced by the laser pulse.

- Geometrical quality of the specimen (flatness and parallelism of the faces), characteristics (homogeneity, isotropy and opacity) and physical properties (thermal expansion vs temperature and sensitivity of the thermal diffusivity to temperature) of the material.

- Operator interventions to select the minimum and maximum of the experimental curve to be normalized, to correct potential thermogram baseline drift and to measure the specimen thickness at room temperature.

- Experimental conditions such as furnace temperature (stability and homogeneity) and nature of the test atmosphere (vacuum, inert gas. . .).

- Differences between the experimental conditions and hypotheses used to establish the theoretical model (finite pulse-time, non-uniformity of the laser beam and thermal losses). Another uncertainty source related to the measurement method is the assumption of the linearity of the infrared detector output with respect to temperature

Among these different factors, the main uncertainty components associated to thermal diffusivity measurement at very high temperature are those related to:

- the "quality" of the experimental curve (noise, drift of baseline during the test, positive or negative shift of the baseline after the laser pulse),
- the differences between the experimental conditions and the assumptions upon which the model has been based,
- the thermal expansion of the specimen during the test if not taken into account in the calculation of the thermal diffusivity.

More details about the assessment of uncertainty associated with high temperature thermal diffusivity measurements can be found in [4].

7. Reference materials

Reference materials with known thermal diffusivities can be used to check the laser flash apparatuses. As there are no reference materials with certified values of thermal diffusivity in the temperature range above 1500 °C to 3000 °C, three refractory materials (molybdenum, tungsten and IG210 isotropic graphite) have been studied as candidate reference materials in the framework of the European project 17IND11 Hi-TRACE.

The thermal diffusivity of these three materials has been measured as a function of temperature by seven laboratories on a batch of 2 mm or 3 mm thick specimens machined in the same blocks of materials. The mean values as well as the standard deviations of reproducibility calculated from the results obtained by the participant laboratories are given in the table 1. The thermal diffusivity values given for temperature higher than 23 °C have been corrected from the thermal expansion of each studied material.

The isotropic graphite can be used up to 3000 °C, while the use of molybdenum and tungsten shall be limited to temperature lower than 2200 °C and 2715 °C respectively due to possible eutectic formation at the surface of the specimens in the case of laser flash apparatus containing graphite parts.

Table 1 Thermal diffusivity of isotropic graphite IG210, molybdenum and tungsten versus temperature

Temperature	Isotropic graphite IG210		Molybdenum		Tungsten	
(°C)	Thermal diffusivity $(10^{-6}\ \mathrm{m^2 \cdot s^{-1}})$	Standard deviation $(10^{-6}\ \mathrm{m^2 \cdot s^{-1}})$	Thermal diffusivity $(10^{-6}\ \mathrm{m^2 \cdot s^{-1}})$	Standard deviation $(10^{-6}\ \mathrm{m^2 \cdot s^{-1}})$	Thermal diffusivity $(10^{-6}\ \mathrm{m^2 \cdot s^{-1}})$	Standard deviation $(10^{-6}\ \mathrm{m^2 \cdot s^{-1}})$
23	84.94	2.31	54.96	1.71	67.65	1.03
50	76.69	2.06	53.90	1.63	66.03	1.02
100	63.79	1.74	51.51	1.51	62.68	1.47
150	54.31	1.46	49.98	1.35	60.11	1.19
200	46.58	0.75	48.07	1.45	57.18	1.28
250	40.77	0.66	46.81	1.31	55.11	1.13
300	36.14	0.73	45.47	1.34	52.83	1.48
400	29.74	0.59	42.89	0.61	49.21	0.98
600	22.30	0.37	39.74	0.43	44.35	0.88
800	18.25	0.30	37.21	0.37	41.14	0.91
1000	15.63	0.28	34.78	0.54	39.12	0.91
1200	13.80	0.23	32.27	0.30	37.05	0.92
1400	12.50	0.17	29.74	0.29	35.42	0.94
1600	11.53	0.15	27.18	0.19	33.77	0.88
1800	10.66	0.20	24.79	0.29	31.91	1.02
2000	9.96	0.14	22.75	0.33	29.89	1.16
2200	9.39	0.15	21.09	0.60	27.79	1.25
2400	8.93	0.19			26.90	1.02
2600	8.44	0.24				
2800	8.11	0.24				
3000	7.64	0.19				

8. References

[1] W.J. Parker, R.J. Jenkins and G.L. Abbott, “Flash method of determining thermal diffusivity, heat capacity, and thermal conductivity”, *J. Appl. Phys.* **32**, 1679 (1961).

[2] T.J. Quinn, Temperature, Academic Press (1990).

[3] G. Failleau, N. Fleurence, O. Beaumont, R. Razouk, J. Hameury and B. Hay, “Metal-carbon eutectic high temperature fixed points for in-situ calibration of radiation thermometers”, *High Temp.-High Press.* **50**, 149 (2021).

[4] B. Hay, O. Beaumont, G. Failleau, N. Fleurence, M. Grelard, R. Razouk, G. Davée and J. Hameury, “Uncertainty assessment for very high temperature thermal diffusivity measurements on molybdenum, tungsten and isotropic graphite”, *Int. J. Thermophys* **43,** 2 (2022).

[5] JCGM 100:2008 (GUM 1995 with minor corrections) - Evaluation of measurement data - Guide to the expression of uncertainty in measurement (2008).