Description of the Laser Flash Analysis Method for Thermal Diffusivity Measurement with the LFA 447

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by

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1. Foreword

The aim of this lecture note is to detail the methodology of the Laser Flash Analysis for the measurement of thermal diffusivity of small test samples. Such measurements can be conducted with the Laser Flash Apparatus LFA 447 (Netzsch Gerätebau GmbH [1]) at the Building Material Characterization Laboratory of Aalborg University - Department of Civil Engineering [2].

2. Introduction

The mathematical equation at the core of most of the heat transfer and thermodynamics models and simulation tools in sciences and engineering is the "Heat Equation":

$$\frac{\partial \theta}{\partial t} = \alpha \nabla^2 \theta \qquad (1)^*$$

Where,
θ: Temperature [K or °C]
t: Time [sec]
α: Thermal diffusivity [m^2/s]

**In here, the heat equation is in its simplest form and only accounts for heat transfer by conduction without internal volumetric heat source, advection or radiative heat transfer terms.*

Solving this differential equation enables to calculate the heat transfer and temperature spatial distribution as a function of time. One can notice that the thermal diffusivity is the only parameter of this equation.

The thermal diffusivity of a material (usually noted α or D and commonly expressed with the SI unit m^2/s or mm^2/s) is related to the effective thermal inertia of a solid and determines how fast heat is propagated through the latter. Knowing this parameter is thus crucial to calculate the thermodynamics, heat transfer and temperature distribution inside a solid over time.

Reciprocally, knowing the transient temperature distribution in a solid, its evolution in time together with the boundary conditions of the experiment, can allow using numerical or analytical solutions of the heat equation to determine the thermal diffusivity of the tested system.

In addition, the measurement of the thermal diffusivity can help to determine other fundamental material characteristics of the studied system:

$$\alpha = \frac{\lambda}{\rho \cdot C_p} \qquad (2)$$

Where,
α: Thermal diffusivity [m^2/s]
λ: Thermal conductivity [W/m.K]
ρ: Density of the material [kg/m^3]
C_p: Specific heat capacity of the material [J/kg.K]

3. History and Description of the Laser Flash Analysis Method

The large interest and the rapid technological developments in the fields of weaponry, aeronautics and aerospace after the Second World War intensified the need for accurate measurement of material thermal properties at high temperatures [3]. Some straightforward steady-state measurement technics, such as the Guarded Hot Plate method or the Heat Flow Meter method [4], enable the measurement of the thermal conductivity of mesoscale test samples. However, they are usually restricted low thermal conductivity samples, are not applicable for high temperature tests, and require a long measurement time. By the end of the 1950s, several non-steady-state measurement methods were available to measure the thermal conductivity and thermal diffusivity of materials but they also presented troublesome limitations: long measurement time, large sample size required, difficult to perform measurements at high temperatures. The main flaw of the classical measurement techniques of that time is that they were not able to maintain adequate boundary conditions during the experiment so that known solutions to the heat equation could be used to calculate the material thermal properties. In particular, some issues were encountered regarding the thermal contact resistance between the tested sample, the heat source, and the heat sink. In addition, minimizing the surface heat losses from the tested sample to its surroundings was also an important challenge. If convective heat losses can be eliminated by conducting the measurement in a vacuum chamber, radiation heat losses become considerable at high temperatures above 1000 °C [3].

In that context, Parker et al. [3] introduced in 1961 a new contactless non-steady-state material thermal characterization technique: the Laser Flash Analysis (LFA). The LFA method enables to rapidly determine thermal diffusivity, specific heat capacity and thermal conductivity of small-size samples of all kinds of solid material at any temperature range. The issues associated with the thermal contact resistance of the heat source are eliminated by using a flash lamp to illuminate and heat up one side of the test sample. The heat losses of the test sample to the surrounding are minimized by the very short measurement time, and considered negligible (adiabatic conditions which are adequate for materials with large thermal diffusivity).

The principle of the Laser Flash Analysis for thermal diffusivity measurement is as follows: One side of the tested sample (here, the bottom side) is illuminated by a high-intensity and short-duration laser flash pulse. This sample's side is thus very rapidly heated up. On the other side of the tested sample (top side), the temperature is measured continuously with a thermocouple or an infra-red sensor (see *Figure 1*). The temperature recording of the top surface starts at the firing of the laser flash and lasts between a few hundred milliseconds and up to a few seconds.

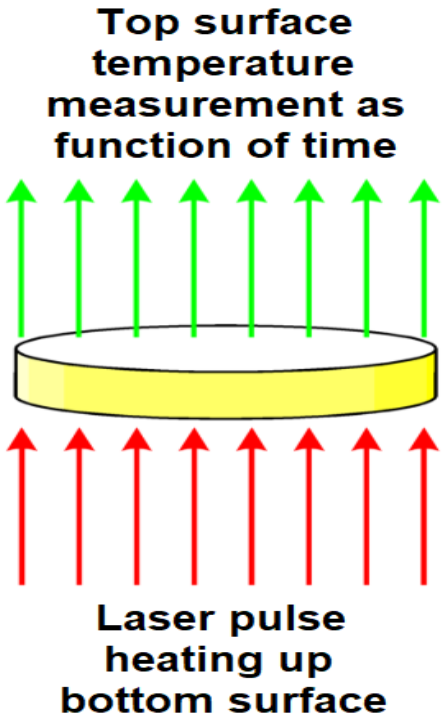

Figure 1: *Principle of the Laser Flash Analysis method.*

One can see in *Figure 2* a typical transient temperature response of a Laser Flash experiment on a thin sample with large thermal diffusivity and for adiabatic conditions with no or negligible heat losses during the temperature record.

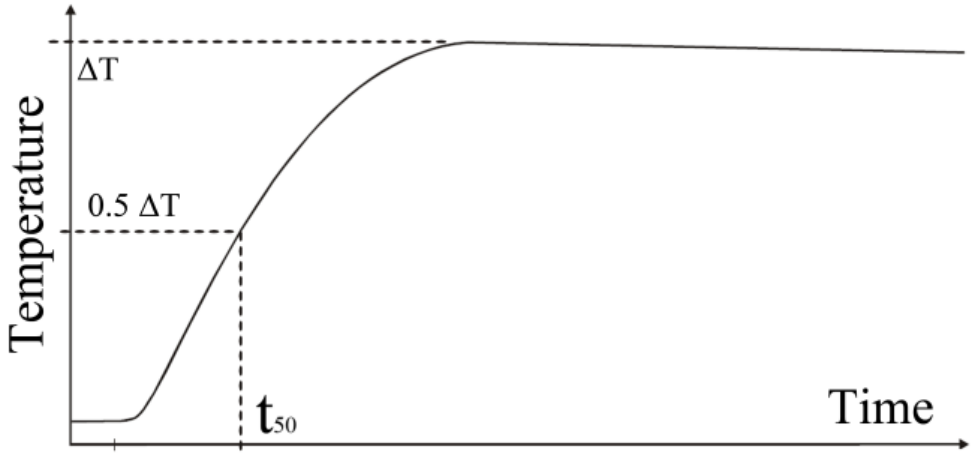

Figure 2: *Typical transient temperature response curve for adiabatic conditions.*

Assuming a one-dimensional heat transfer inside a perfectly thermally insulated solid of uniform thickness L, and given an initial temperature distribution $\theta(x,0)$ within that solid, the temperature distribution inside the latter can be calculated with the following equation presented by Carslaw and Jaeger [5]:

$$\theta(x,t) = \frac{1}{L}\int_0^L \theta(x,0)dx + \frac{2}{L}\sum_{n=1}^{\infty} exp\left(\frac{-n^2\pi^2\alpha t}{L^2}\right) \times cos\frac{n\pi x}{L}\int_0^L \theta(x,0)cos\frac{n\pi x}{L}dx \quad (3)$$

Where x is the distance inside the sample from the bottom side, t is the time elapsed from the laser flash firing, L is the uniform thickness of the sample, and α is the thermal diffusivity of the tested sample's material.

Based on the aforementioned equation (3) and assuming that the tested sample is subjected to one-dimensional heat transfer only with adiabatic boundary conditions without any heat losses to the ambient, and that the short laser flash pulse is instantaneously and uniformly absorbed at the bottom surface of the sample over a very small depth (which is the case for opaque materials and laser flash test samples which are coated with a thin absorbent graphite layer), Parket et al. [3] developed an analytical solution to the heat equation modelling the results (transient temperature response at the top surface of the test sample) of the Laser Flash experiment:

$$\theta(t) = \frac{Q}{\rho C_p L}\left[1 + 2\sum_{n=1}^{\infty}(-1)^n exp\left(\frac{-n^2\pi^2}{L^2}\alpha t\right)\right] \qquad (4)$$

Where Q is the energy density of the laser flash pulse which is instantaneously and uniformly absorbed by the bottom surface of the sample, ρ is the sample's material density, C_p is the sample's specific heat capacity, L is the uniform thickness of the sample, and α is the thermal diffusivity of the tested sample's material.

This model can be simplified. The density and specific heat capacity terms can be eliminated by introducing a new term θ_{max} accounting for the maximum temperature occurring on the top surface of the sample during the laser flash experiment, which yields the following equation modelling the temperature history of the top surface:

$$\theta(t) = \theta_{max}\left[1 + 2\sum_{n=1}^{\infty}(-1)^n exp\left(\frac{-n^2\pi^2}{L^2}\alpha t\right)\right] \qquad (5)$$

From this analytical solution, Parker et al. then deduced a simple formula to calculate the thermal diffusivity α of the sample's material with only two experimental parameters which are easy to evaluate: the thickness of the sample L, and the half-time t_{50} at which the temperature rise of on the top surface of the sample reaches half of its maximum value; $(\theta_{max} - \theta(0))/2$ (see *Figure 2*):

$$\alpha = \frac{0.1388 \times L}{t_{50}} \qquad (6)$$

Although the calculation method developed by Parker et al. is very easy to use, it is restricted to near-adiabatic experimental conditions which can only be met for high thermal diffusivity samples with small thickness and short measurement period (around 100 milliseconds or lower). However, for samples with lower thermal diffusivity, the measurement duration is too large for heat losses by convection and radiation to be neglected. For high-temperature measurements (above 1000 K), radiation heat losses between the tested sample and the surrounding are especially predominant and cannot be neglected for LFA thermal diffusivity calculation.

In 1963, shortly after the publication of Parker et al., different numerical models, such as Cowan's model [6] and Cape and Lehman's model [7], have been developed to account for heat losses by radiation and convection at the sample's surfaces during the Laser Flash experiment. They thus enable the analysis of Laser Flash experimental data for lower-thermal conductivity samples and determine correctly the thermal properties of nearly all types of materials at low and high temperatures above 2500 K [6].

In addition, the numerical model of Parker et al. assumes that the duration of the laser flash pulse is infinitely short and that the energy of the former is instantaneously transferred to the bottom surface of the sample. This is an adequate assumption if the duration of the laser pulse is very small and negligible in comparison to the time it takes for heat to diffuse through the solid. If this is not the case, the laser pulse duration can be taken into account by certain models such as the ones presented by Cape and Lehman [7] and Larson and Koyama [8].

Another phenomenon which should be taken into account when treating Laser Flash Analysis data is the initial direct radiative heat transfer (also denominated as "ballistic transport") inside the test sample. This is characterized by a quasi-immediate temperature measurement peak and plateau right after the firing of the laser flash (at time = 0), before the temperature signal increases again due to diffusion heat transfer in the solid (see *Figure 3*). This phenomenon occurs in certain materials are partially or fully transparent in the near-infrared, such as sedimentary minerals quartz and feldspar [9]. A rapid heat transfer by radiation from the bottom surface (illuminated by the laser pulse) to the top surface of the sample thus appears almost immediately after the laser pulse. This initial ballistic heat transport is not related to the diffusion heat transfer by conduction in the solid, but only due to rapid long-wave (near-infrared) radiation heat transfer occurring at the speed of light. This ballistic radiative heat transfer should, therefore, be corrected or it will lead to an overestimation of the material thermal diffusivity.

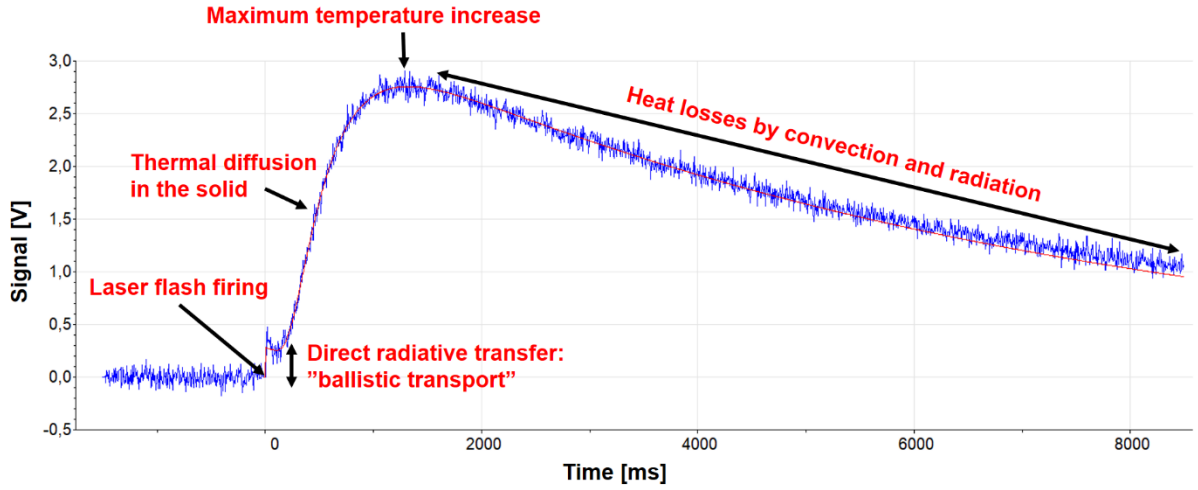

Figure 3: *Transient temperature response curve of Laser Flash experiment with initial direct radiative heat transfer (ballistic transport) followed by thermal diffusion in the solid with heat losses by convection and radiation from the sample's surfaces to the environment.*

Cowan's model and Cape and Lehman's model do not account for this initial direct ballistic radiative heat transfer. Consequently, Mehling et al. [10] suggested in 1998 a new frequency-dependent absorption model accounting for the latter, allowing for correct thermal diffusivity calculation from the Laser Flash Analysis data. This model is denominated as "Radiation model" in the LFA data treatment software [1].

4. Laser Flash Analysis for Thermal Diffusivity Measurement with the LFA 447 Apparatus

Nowadays, the Laser Flash Analysis is a widely used state-of-the-art transient measurement method for the determination of the thermal diffusivity of materials. It can be used with all types of materials (including powders, liquids and slurries), as long as they can be arranged as a thin sample of uniform thickness with even and parallel surfaces which can be coated with an absorbent layer such as graphite spray.

The Laser Flash Analysis apparatus used for thermal diffusivity measurement at the Building Material Characterization Laboratory of Aalborg University - Department of Civil Engineering [2] is an LFA 447 from Netzsch Gerätebau GmbH [1]. This LFA 447 is equipped with a matrix mode for local measurement of thermal properties on large samples. This can be very convenient for the study of large graded material samples with continuous change of thermal properties along the plane of the sample [11]. The apparatus can load up to 4 samples for automated measurement. The laser flash source is a xenon lamp is placed underneath the sample holder. The infrared detector is located above the sample changer with the liquid nitrogen tank (for infrared sensor cooling) above it (see *Figure 4*). The temperature of the sample holder is controlled by a furnace and a water-based cooling system. The sample's temperature can thus be set between 10 °C and 300 °C.

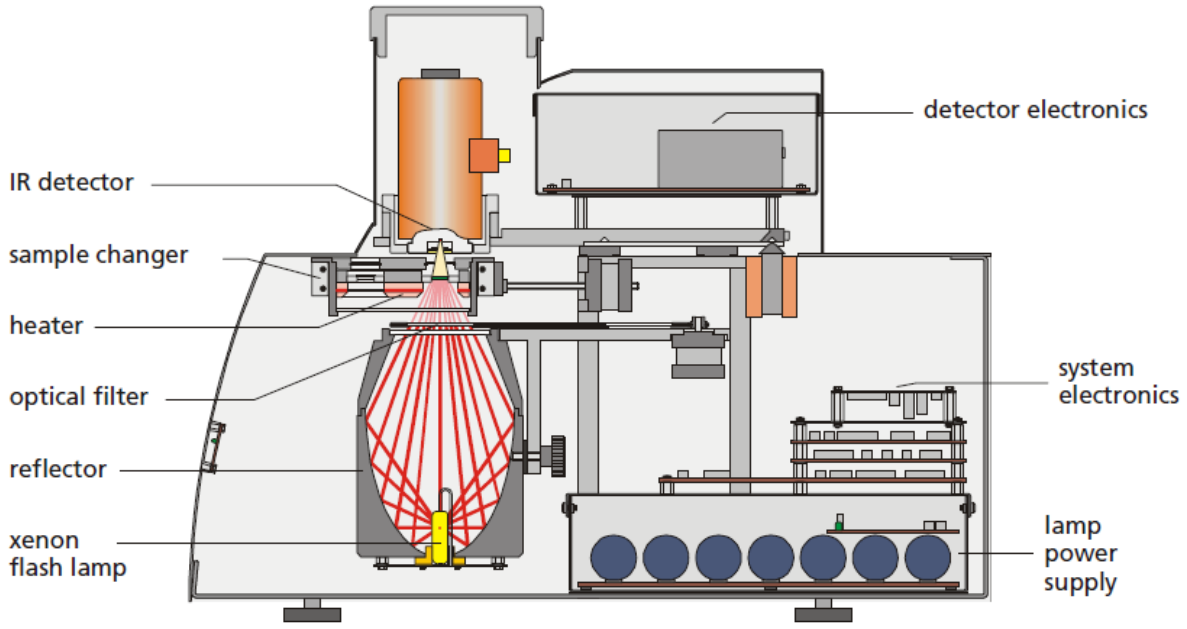

Figure 4: *Overview of the Laser Flash Analysis apparatus LFA 447 from Netzsch Gerätebau GmbH [1].*

For the LFA 447, the optimum thickness of the test sample is between 1 mm and 3 mm and should be chosen according to the technical recommendations (see *Table 1*). The LFA experiment should be conducted with a measurement duration ranging from 10 ms to 20 000 ms.

Table 1: *Suggested test sample's thickness as a function of material thermal diffusivity [1].*

Diffusivity	**Suggested thickness** (mm)
low diffusivity e.g. polymers (0.001cm^2/s)	1 to 1.5
medium diffusivity e.g. ceramics (0.05 cm^2/s)	1.5 to 2
high diffusivity e.g. copper (1 cm^2/s)	2 to 3

During the raw data treatment, the operator should make sure that the LFA measurement records are meaningful with a temperature history curve resembling one of the possible scenarios presented in *Figure 5*. The operator should then select the model which fits the best temperature history curve of the LFA experiment. From the selected model, the thermal diffusivity of the test sample's material can be calculated directly by the Netzsch Proteus® LFA Analysis software performing the experimental data treatment [1].

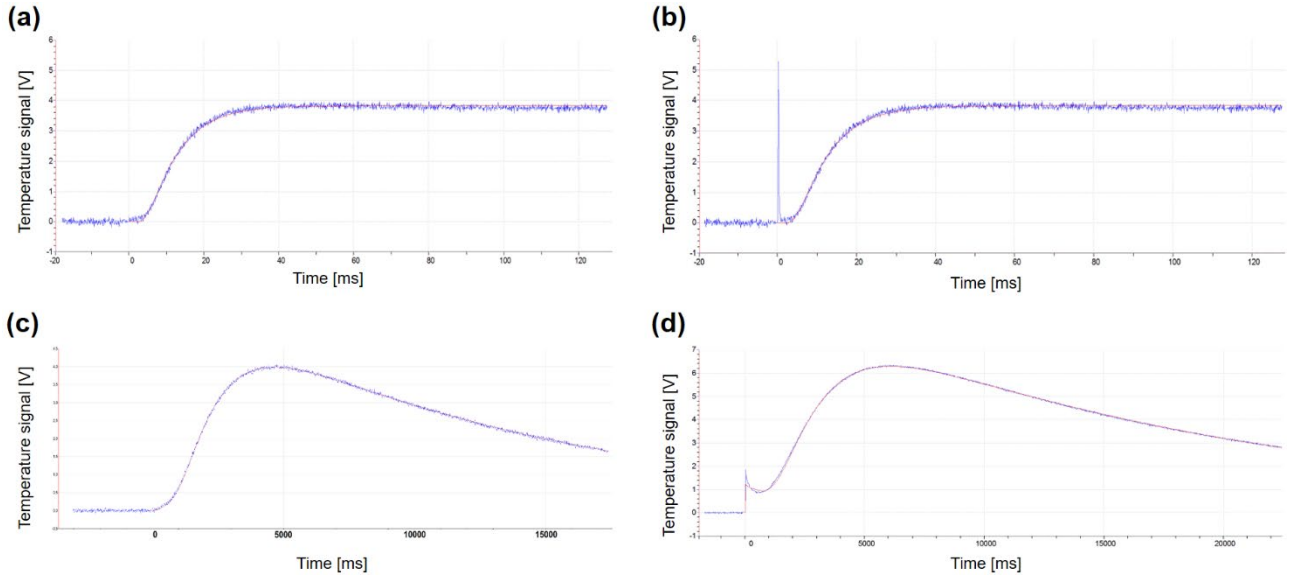

Figure 5: *Typical experimental raw data from Laser Flash Analysis: (a) adiabatic conditions for highly conductive sample without heat losses; (b) adiabatic conditions for highly conductive sample without heat losses and with initial temperature signal peak caused by internal reflection in the machine of the laser pulse reaching directly the infra-red detector; (c) non-adiabatic conditions for low thermal diffusivity sample with heat losses; (d) non-adiabatic conditions for low thermal diffusivity sample with heat losses and initial ballistic heat transfer by radiation inside the near infra-red transparent sample.*

The Laser Flash Analysis method can also be used to measure the specific heat capacity of a sample's material and thus calculate its thermal conductivity [1]. However, it is recommended to use a Differential Scanning Calorimetry (DSC) method instead, if possible.

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