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A new laser flash system for measurement of the thermophysical properties

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Abstract

The flash method is a well-known technique for measurement of the thermophysical properties (thermal diffusivity, specific heat and thermal conductivity) of solid materials. Easy sample preparation, fast measurement times and high accuracy are only some of the advantages of this non-contact, non-destructive testing technique.

A new laser flash system, the NETZSCH LFA 457 MicroFlash, was developed for measurement of the thermophysical properties (thermal diffusivity, specific heat, thermal conductivity). The system can be equipped with two different user-interchangeable furnaces allowing measurement between −125 and 1100 °C. The vacuum-tight construction of the system allows tests under well-defined atmospheres as well as under vacuum. The integrated automatic sample changer enables testing of up to three samples at the same time. Presented in this work are the technical details of the instrument and some performance tests as well as various application examples on industrially relevant materials.

Keywords: Laser flash; Thermal conductivity; Specific heat; Thermal diffusivity; Solids; Composites

1. Introduction

Over the past few decades, the flash method [1] has developed into one of the most widely used techniques for measuring the thermal diffusivity and thermal conductivity of various kinds of solids, powders and liquids. This technique entails heating the front side of a small, usually disk-shaped plane-parallel sample by a short energy (laser) pulse. The temperature rise on the rear surface is measured versus time using an infrared detector. By placing the sample into a tube furnace, temperature-dependent measurements can easily be carried out as well.

Since introduction of the method by Parker et al. [1], various improvements to it have been made. New evaluations have been developed which account for heat loss [2] and finite-pulse effects [3]. Some analysis methods are already available which simultaneously take both of these into account in the optimum way [4]. Furthermore, modern systems allow determination of the thermal diffusivity a and the specific heat c_p. Once the bulk density ρ of the material is known, a direct thermal conductivity determination is possible according to Eq. (1):

$$\lambda(T) = \rho(T)c_p(T)a(T) \quad (1)$$

The new NETZSCH model LFA 457 MicroFlash combines modern technology with state-of-the-art data processing techniques [5]. The fully automated top loading flash system with an Nd:YAG laser on the bottom and an infrared detector on top allows for easy sample loading and flexible sample geometries while providing an optimum signal-to-noise ratio. The specially designed 500 kHz data acquisition system allows measurement of critical samples such as thin films or high-conductivity materials. The system is fully software-controlled.

The measured data is analyzed taking both heat loss and finite-pulse effects into consideration. Standard constituents of the analysis software are the improved Cape-Lehmann model [5], the possibility of considering for radiative heat transfer, and non-linear regression routines for the characterization of two- and three-layer systems.

2. Experimental

Presented in Fig. 1 is the schematic design of the NETZSCH LFA 457 MicroFlash (measurement part). Positioned in the base

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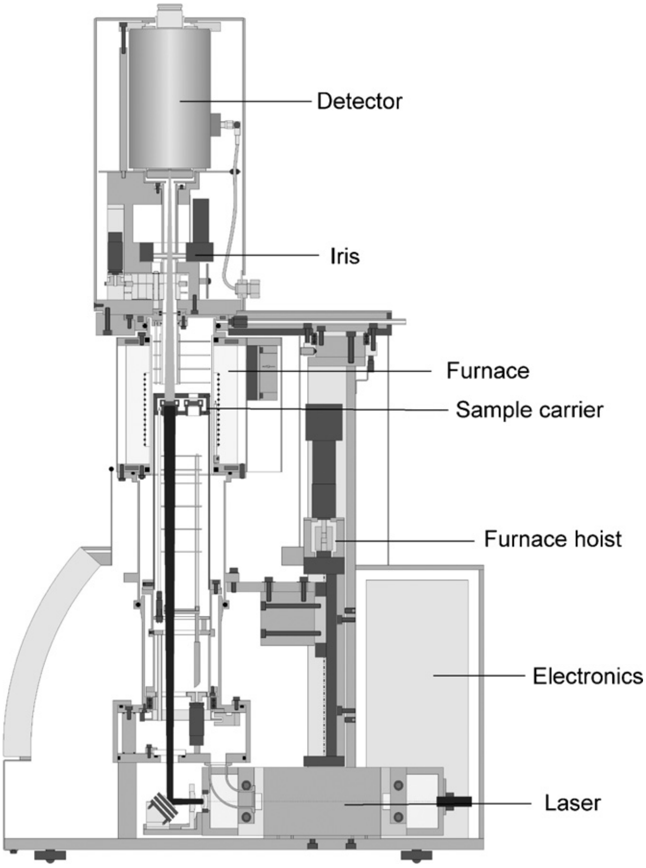

Fig. 1. Measurement part of the NETZSCH LFA 457 MicroFlash.

of the device is the head of an Nd:YAG laser. The laser has a pulse length of 330 μs and a pulse energy output of up to 15 J/pulse. Power is supplied to the laser by a capacitor bank positioned in a separate box. The power output of the laser can be controlled by the software via the voltage level of the capacitor bank and/or via a filter system positioned in the outlet area of the laser system. The laser pulse is deployed through an enlargement optics system which adjusts the beam diameter to the required sample diameters. From the enlargement optics system, the laser pulse is guided via a mirror through a window into the vacuum-tight sample chamber.

Inside the sample chamber is an automatic sample changer for up to three samples. The samples are positioned in easily user-interchangeable sample carriers which can be adjusted to the actual sample dimensions (square samples, disk-shaped samples with various diameters, etc.). A detailed schematic of the sample holder arrangement is presented in Fig. 2. By employing special sample carriers, the system can be used to analyze liquids or metals through the melt. The sample carriers sit on a sample holder tube which rotates to bring the required sample into the measurement position. The entire sample holder system is surrounded by a user-interchangeable furnace. Two furnaces (−125–500 °C or 25–1100 °C) can be used on the system. The temperature rise on the back surface of the sample is measured employing either an InSb- or an MCT-infrared detector. Data is acquired via high-speed amplifier and A/D-converter systems with a maximum possible data acquisition rate of 500 kHz.

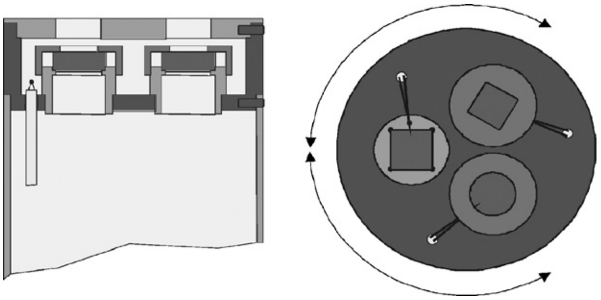

Fig. 2. Schematic design of the sample carrier system (side view and top view) of the LFA 457 MicroFlash.

System control and evaluation of the measurement results are carried out using a 32-bit MS® Windows™ software which allows fully automatic tests and provides state-of-the-art analysis routines for the processed data. The software package includes non-linear regression routines to consider for radial and facial heat losses, finite-pulse effects, and internal radiative heat transfer. Two- and three-layer systems can also be analyzed. The specific heat can be determined and therefore, the thermal conductivity can be directly measured as well. Tests on standard materials such as NIST certified reference materials or pure metals have demonstrated that the system has an accuracy of generally better than 3%. By employing special container systems (made of sapphire, aluminium or platinum), the system can also be used for liquids. Such containers can simply be placed on the sample carrier plate and the tests can be carried out as they normally would [6].

3. Results and discussion

Shown in Fig. 3 are the thermal diffusivity results on copper and aluminium between room temperature and 600 °C, together with literature values [7]. The literature values taken have a stated uncertainty of 3–4%. Comparing the measurement results with the literature values, it can be seen that the deviations between the results are less than the uncertainty of the literature values.

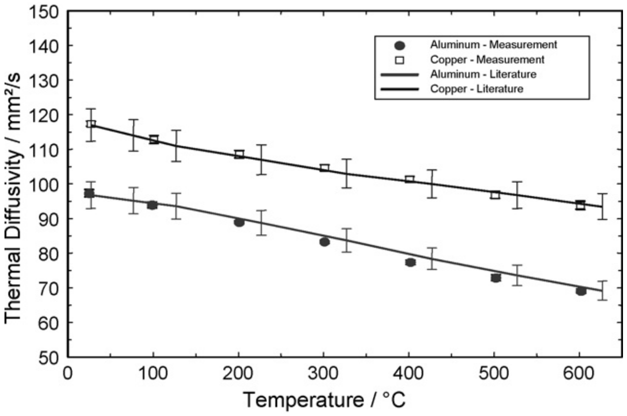

Fig. 3. Thermal diffusivity of copper and aluminium between room temperature and 600 °C (measurement results and literature values).

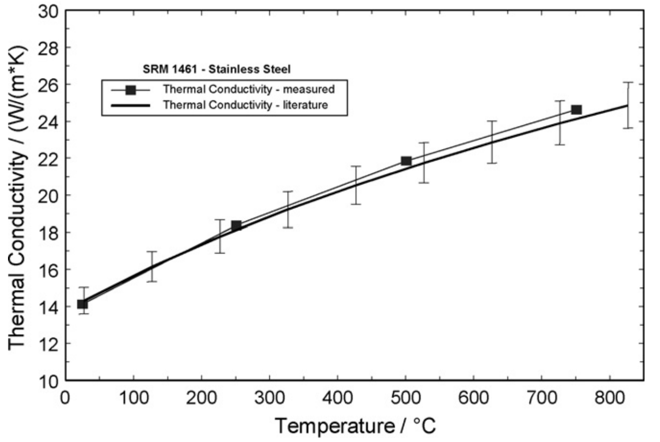

Fig. 4. Thermal conductivity of NIST SRM 1461 stainless steel (comparison between measurement results and the values from the certificate).

Fig. 4 shows the result of a direct thermal conductivity determination (measurement of room temperature bulk density, specific heat and thermal diffusivity and calculation of the thermal conductivity) of an NIST certified thermal conductivity standard SRM 1461 stainless steel. The test results are compared with the values listed in the NIST certificate [8]. As can be seen, the deviations between the test results (symbols) and the values from the certificate (line with error bars) are generally within approx. 3%. This is within the stated uncertainty of this standard reference material.

Presented in Fig. 5 are the measurement results of an NR/BR rubber mixture between −125 and 75 °C. The specific heat used for calculating of the thermal conductivity was measured by an additional DSC test. It can clearly be seen that the thermal diffusivity decreases over the entire temperature range. Between −75 and −50 °C, a step is visible in the thermal diffusivity. The specific heat increases over the entire temperature range. Also here, a step is visible in the same temperature range as the step in the thermal diffusivity results. Both steps can be explained by a glass transition in the rubber material. The thermal conductivity does not show any effects of the glass transition; a nearly linear increase was obtained over the entire temperature range.

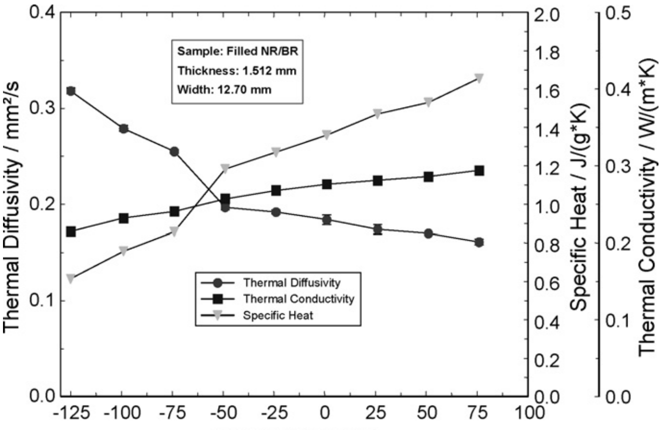

Fig. 5. Specific heat, thermal diffusivity and thermal conductivity of an NR/BR rubber mixture between −125 and 75 °C.

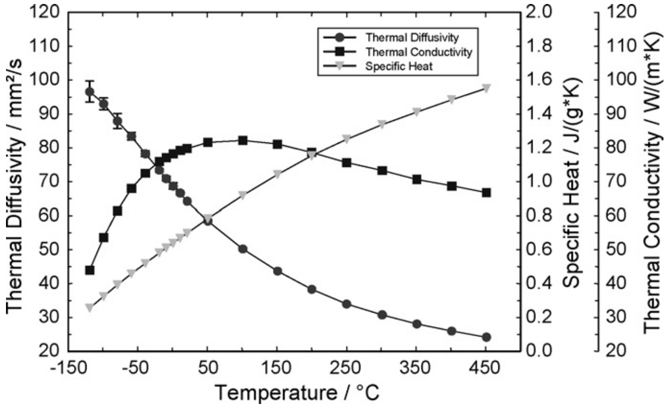

Fig. 6. Specific heat, thermal diffusivity and thermal conductivity of a polycrystalline graphite material between −125 and 450 °C.

Presented in Fig. 6 are the specific heat, thermal diffusivity and thermal conductivity of a polycrystalline graphite material. Such materials are quite interesting for several reasons: the density does only shows a weak dependence versus temperature [9]. Also the heat transfer is mainly based on the contribution of the lattice structure. The specific heat, however, shows a large increase versus temperature below room temperature. This can be explained by the Debye theory [10], which describes the temperature dependence of the specific heat. At low temperatures, the specific heat should increase proportionally to T^3 (temperature in Kelvin). At high temperatures, the specific heat should converge to a constant value. The temperature range in which the material starts to differ from the T^3 depends on the Debye temperature, which is a material-specific property. This temperature is comparably high for carbon materials (above 2000 K). Therefore, one can expect that the specific heat below and around room temperature still closely resembles the T^3 behavior. The thermal diffusivity depends mainly on the mean free path length of the phonons. This mean free path length decreases versus temperature. Therefore, the thermal diffusivity should be proportional to T^{-1} [11].

Determining the thermal conductivity according to Eq. (1) should therefore show an increase versus temperature proportional to T^2 at low temperatures and a decrease proportional to T^{-1} at high temperatures. Considering such a temperature dependence, a maximum should occur in the thermal conductivity in the range between. For most ceramic materials, this maximum is generally in the low-temperature range. Due to the high Debye temperature of graphite, however, its maximum should be around or slightly above room temperature. As can be seen from Fig. 6, the test results follow the expected theoretical behavior almost exactly. Only at temperatures below −50 °C is the thermal diffusivity slightly below the expected temperature dependence. However, one should keep in mind that the mean free path length of the phonons cannot increase infinitely when the temperature is decreasing. The mean free path length cannot be longer than the size of a crystallite inside the sample. This, of course, results in the fact that the thermal diffusivity cannot increase infinitely but should converge to a constant value which is dependent on the structure of the material itself. This

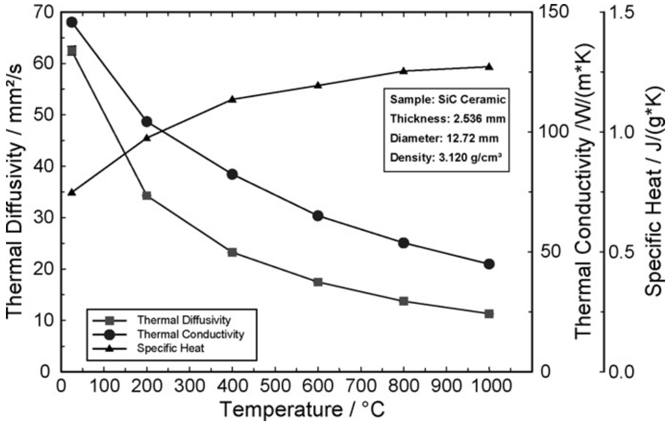

Fig. 7. Specific heat, thermal diffusivity and thermal conductivity of a silicon carbide (SiC) ceramic between room temperature and 1000 °C.

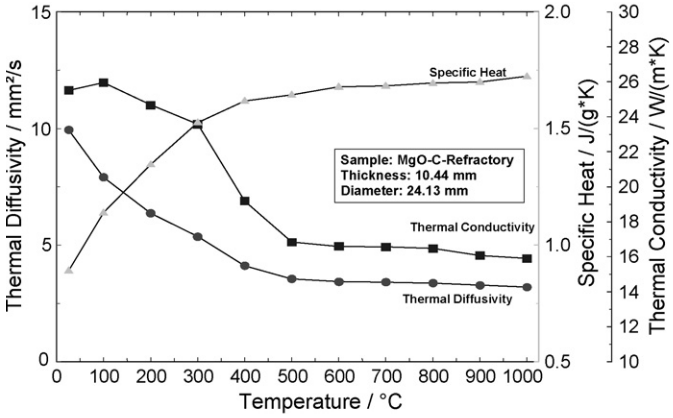

Fig. 8. Thermophysical properties of a magnesia carbon refractory material (20 wt.% carbon content).

explains the low-temperature behavior of the thermal diffusivity. The thermal conductivity shows the expected maximum which was measured around 75 °C.

Depicted in Fig. 7 are the thermophysical properties of a SiC (silicon carbide) ceramic between room temperature and 1000 °C. The thermal diffusivity and specific heat were determined in the LFA 457. Using the measured data, the thermal conductivity was calculated by multiplying the thermal diffusivity, specific heat and room-temperature bulk density. The thermal diffusivity values decrease over the entire temperature range. The specific heat increases, as can be expected from the Debye theory. The thermal conductivity decreases over the entire temperature range, as well. However, it is at a very high level [nearly 150 W/(m K)] at room temperature, which is typical for polycrystalline SiC ceramics.

Presented in Fig. 8 are the test results (thermal diffusivity, specific heat and thermal conductivity) measured on a magnesia–carbon refractory material with a carbon content of approx. 20%. The specific heat increases versus temperature as expected from the Debye theory. Furthermore, the results are between the typical values for pure magnesia and pure carbon [12]. The thermal diffusivity decreases versus temperature over the entire temperature range. Between 300 and 500 °C, a step was detected in the results which can be explained by the decomposition of the organic binder. The decomposition of the binder yields a slightly higher porosity and carbon black between the grains of the refractory. This results in an increased thermal resistance between the grains, causing a decrease in the thermal diffusivity of the entire material. The thermal conductivity shows a slight increase up to 100 °C, which can be explained by the strong increase in the specific heat in this temperature range. This increase compensates for the decrease in the thermal diffusivity. At higher temperatures, the changes in the thermal conductivity are mainly controlled by changes in the thermal diffusivity. The slight drop in the thermal conductivity above 800 °C is most probably due to contact/surface reactions between the carbon and the oxide material inside the refractory. The value changes from ∼25 W m^{-1} K^{-1} at room temperature to ∼16 W m^{-1} K^{-1} at 1000 °C, typical for magnesia refractories with a significant carbon content.

4. Conclusion

A new laser flash device was developed for the characterization of a wide range of materials in solid and liquid state. Tests on standard materials have proven the reliability of the new system. The uncertainty for the thermal diffusivity is 3% for most materials and sample sizes. The uncertainty for the specific heat and therefore for a ‘direct’ thermal conductivity determination is 5–7%. The measurement results presented on a wide range of materials demonstrate the capability of the instrument in various application fields.

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