Investigation of the influence of technological factors on the uncertainty of the results of measuring thermal conductivity by the method of laser flash

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The estimation of the deviation in the measurements of thermal conductivity by the laser flash method for materials with different thermal conductivity coefficients, arising due to the presence of a graphite coating on the sample and the small thickness of the sample, is carried out. A computer model of the method was created in the Comsol Multiphysics software environment. For bulk samples with a graphite coating thickness of $20\,\mu$m, the deviation is 5.5%. The thickness of bulk samples does not affect the measurement results. For materials with low thermal conductivity, a sharp increase in the deviation is observed, reaching 60%. For thermally conductive materials, the deviation is 16−18%. For thin samples less than $10\,\mu$m thick, the thickness of the graphite coating does not affect the measurement results. The decisive factor is the duration of the laser pulse.

Keywords: laser flash method, thermal conductivity, relative deviation.

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1. Introduction

The laser flash method belongs to non-stationary methods for measuring thermal conductivity. It is widely used to measure the thermal conductivity of various materials such as ceramics, metals, semiconductors, thermoelectric materials and other functional materials [1–4]. To do this, one side of the sample is irradiated with a short laser pulse, and then the temperature change is recorded on the other side [5]. Using the obtained measurement results, knowing the values of density (ρ) and specific heat capacity (c_P) of the material, it is possible to determine the thermal conductivity coefficient (λ) of the sample by the formula

$$\lambda = 1.36976 \cdot \rho \cdot c_p \cdot \frac{h_0^2}{\pi^2 \tau_{1/2}}, \tag{1}$$

where h_0 is the sample height, $\tau_{1/2}$ is the time to reach half the maximum temperature on the reverse side of the sample.

There are a number of recommendations and limitations for this method, such as the time finitude of the laser pulse [6], uneven heating of the sample, thickness, shape and opacity of the sample [7,8]. In most experiments, to ensure the greatest absorption of pulse energy by the sample and increased accuracy of the temperature rise measurements, the sample is covered with a layer of graphite.

2. Creating a computer model

To assess the effect of various factors on the deviation of the measured thermal conductivity coefficient from the true value, several mathematical models were created that describe the effect of each of these technological factors on the measurement process. The modeling was carried out in the Comsol Multiphysics software environment.

The geometric model of the studied sample is presented in the form of a graphite-coated cylinder (Fig. 1). The computer model considered a homogeneous isotropic sample with a fixed value of the thermal conductivity coefficient, diameter $d = 30$ mm, and height $h_0 = 2$ mm. The graphite layer with thickness h_1 covered the entire sample surface. The underside of the sample was irradiated with a radiant energy pulse having a time distribution in the form of a Gaussian. The measurements were modeled in an atmosphere of blowing with gaseous nitrogen.

The energy flux from the laser pulse propagates in the sample volume according to the Fourier law:

$$\mathbf{q} = -\lambda \nabla T, \tag{2}$$

which leads to temperature increasing on the opposite side of the sample. Here $\mathbf{q}$ is the heat flux density vector, T is the temperature.

The temperature distribution in a given region of space and its change in time is described by the basic equation of heat conductivity:

$$\rho c_p \frac{\partial T}{\partial t} + \nabla \cdot \mathbf{q} = 0. \tag{3}$$

Boundary conditions for the irradiated surface:

$$-\mathbf{n} \cdot \mathbf{q} = q_b, \tag{4}$$

where $\mathbf{n}$ is the normal vector to the heat exchange surface, q_b is the heat flux density from the radiant energy pulse. For the remaining surfaces the boundary condition for radiative heat exchange and convection heat exchange was used:

$$-\mathbf{n} \cdot \mathbf{q} = \varepsilon\sigma(T_{amb}^4 - T^4), \tag{5}$$

$$q_0 = \alpha \cdot (T_{ext} - T). \tag{6}$$

Here σ is Stefan–Boltzmann constant, ε is body emissivity factor, T_{amb} is ambient temperature, T_{ext} is nitrogen gas temperature. ε value was taken equal to 0.9 for all surfaces of samples. T_{amb} value was taken to be 293.15 K.

For horizontal plates with the heat transfer surface facing upwards, the heat transfer coefficient α can be calculated as in [9]:

$$\alpha = 1.3 \cdot \frac{Nu \cdot \lambda_f}{l_0}, \tag{7}$$

$$Nu = C \cdot Ra^m, \tag{8}$$

where Ra is Rayleigh number, which is determined by thermophysical properties of gaseous nitrogen, temperature difference and defining size, and for this model $Ra = 5469$. C and m are coefficients determined depending on the mode of motion of the environment (for our case $C = 0.54$ and $m = 0.25$), λ_f is thermal conductivity coefficient of gaseous nitrogen at an average temperature of the medium, l_0 is the determining size (for disk the determining size is its diameter d).

Based on the initial data, the calculated value of the heat transfer coefficient was $\alpha = 5.1\,\mathrm{W/(m^2 \cdot K)}$.

In the model, the duration of the laser pulse was set as $t_{imp} = 2$ ms, which corresponds to the factory parameters of NETZSCH LFA 457 MicroFlash unit. The pulse power was chosen such that the temperature change on the underside of the sample did not exceed 2 K.

On the basis of mathematical modeling, the dependences of the temperature increasing on the underside of the sample on time were obtained (Fig. 2). The expected temperature decreasing due to heat transfer to the environment occurs much later than the simulated time interval and, accordingly, is not shown on the graph. These data were further used to determine the value ($\tau_{1/2}$), which is

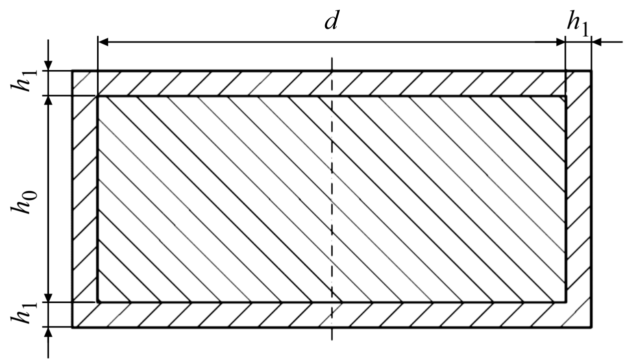

Figure 1. Geometric shape of the sample in the model: h_0 is sample height, h_1 is graphite coating thickness, d is sample diameter.

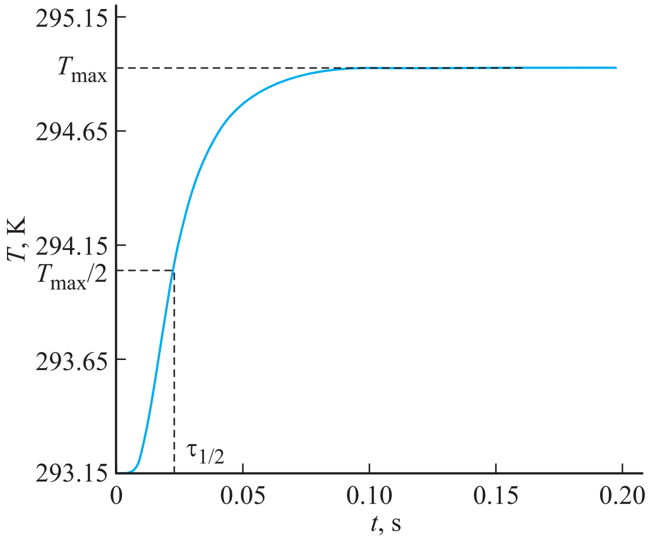

Figure 2. Temperature of underside of the sample vs. time, obtained using computer simulation.

necessary for calculating the thermal conductivity coefficient (λ_c) using formula (1). The obtained calculated value of the thermal conductivity coefficient was compared with the true coefficient (λ_t) specified at the modeling stage. Thus, the effect of the considered factors on the measurement accuracy was assessed.

To visually show the differences between the true and calculated values of the thermal conductivity coefficients, their difference was determined, and scaling according to the true value was made:

$$\delta = \frac{\lambda_t - \lambda_c}{\lambda_t} \cdot 100\%. \tag{9}$$

3. Results and discussion

3.1. Bulk samples

In the paper [10] a study was carried out, and it was found that the thickness of the graphite coating ranges from 5 to 15 μm on one side of the deposition on the sample. When studying bulk samples, the effect of this layer on the result of the measurement process is usually neglected. However, the graphite layer may contribute to the evaluation of measurements. Therefore, we considered the effect of the graphite coating thickness, as well as the thickness effect of the sample itself on the results of the samples thermal conductivity measurement. The samples under consideration had a fixed thermal conductivity $\lambda = 3.4\,\mathrm{W/(m \cdot K)}$.

3.1.1. Effect of graphite coating thickness. The height of the samples of the first series was $h_0 = 2$ mm, and their surface was covered with a graphite layer of different thicknesses h_1. The modeling results are shown in Fig. 3, *a*. Based on these results it was found that with the thickness

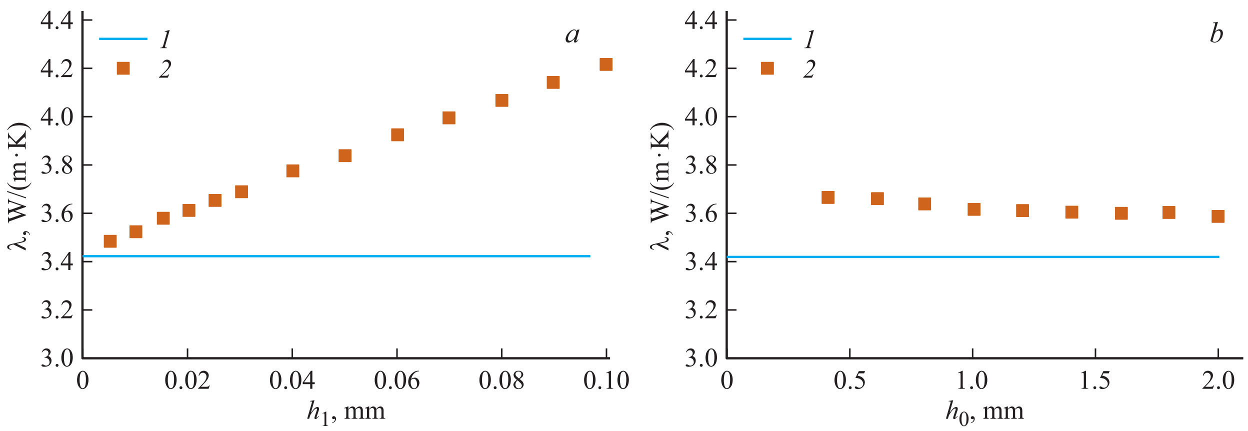

Figure 3. Thermal conductivity coefficient vs. thickness of the graphite coating h_1 (a); sample height h_0 (b). *1* is true value of λ_t, *2* is calculated value of λ_c.

of the graphite coating increasing, a linear increasing of the calculated value of thermal conductivity is observed. With the graphite layer thickness of $100\,\mu$m the deviation δ of the calculated value from the true value reaches 20%. For the graphite coating thickness of $20\,\mu$m, which is close to the actual graphite coating thickness in field experiments, the deviation is 5.5%. In further studies the average thickness of the graphite coating $h_1 = 20\,\mu$m was used.

This behavior of the calculated value of the thermal conductivity coefficient is associated with a high (by more than 30 times) thermal conductivity of graphite.

3.1.2. Effect of sample thickness. In the second series of numerical experiments the sample height h_0 varied in the range of 0.4 to 2.0 mm. The thickness of the graphite coating layer was taken to be constant and equal to $20\,\mu$m. The modeling results are shown in Fig. 3, *b*. Based on the data obtained it can be concluded that the sample thickness has practically no effect on the measurement results. The deviation increasing is observed only at sample thicknesses below 0.7 mm, which is the lower value of the recommended sample thickness for NETZSCH LFA 457 MicroFlash unit.

It can be seen from formula (1) that the calculated value of thermal conductivity is directly proportional to the square of the sample thickness. With the graphite coating increasing, the total thickness of the sample increases, which is not taken into account in the calculation formula leading to the uncertainty increasing in Fig. 3, *a*.

3.1.3. Effect of sample material. The effect of the graphite coating on the measurement results was evaluated for materials with different values of the thermal conductivity coefficient. Materials with thermal conductivity from 0.15 W/(m · K) (polyimide) to 400 W/(m · K) (copper) were considered. The properties of the substances used in the modeling are presented in the Table. The samples had the same height $h_0 = 2$ mm and thickness of the graphite coating layer $h_1 = 20\,\mu$m.

The modeling results are shown in Fig. 4. The modeling showed that for materials with the thermal conductivity

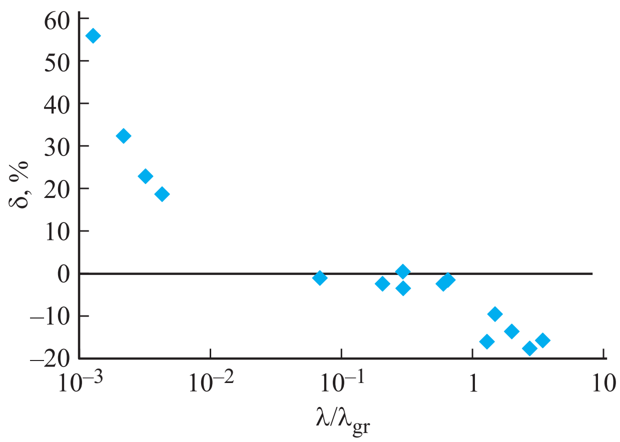

Figure 4. Normalized deviation of measurements of thermal conductivity coefficient λ of bulk samples vs. properties of the material.

coefficient of $0.01\lambda_{gr}$ and below, a sharp increasing of δ is observed, which reaches 60% for polyimide with a thermal conductivity of 0.15 W/(m · K). This dependence can be explained by the presence of a thin layer of graphite on the side surface of the sample, which in this case plays the role of a thermal bridge. In the range from 0.1 to 1 the δ deviation is minimum and comparable to the declared accuracy of the measuring unit. For materials whose thermal conductivity exceeds that of graphite, the increasing of δ with its sign change is observed, since the graphite layer behaves as a heat insulator in such cases.

3.2. Thin samples

The thin samples were individually considered, they do not belong to thin-film structures, but their thickness is lower than thickness recommended by the manufacturer of the measuring equipment (10–100 μm). At sample thickness of several tens of micrometers, the presence of

Properties of substances used in the computer model

	c_P, J/(kg · K)	ρ, kg/m^3	λ, W/(m · K)
Polyimide	1100	1300	0.15
Nylon	1700	1150	0.26
Polyethylene	1900	930	0.38
Mica	880	2900	0.5
Bismuth	122.08	9800	7.8
Antimony	207	6684	24.3
Aluminum oxide	730	3965	35
Lead	127	11340	35.3
Platinum	133	21450	71.6
Iron	440	7870	76.2
Tungsten	132	19350	174
Aluminium	894	2700	236
Gold	129	19300	317
Copper	385	8940	400

the graphite coating can have significant effect on the experimental results. To evaluate them, a numerical modeling of the laser flash method was carried out for samples with thickness of 10 to 70 μm. The samples under consideration had a fixed thermal conductivity $\lambda = 0.15$ W/(m · K). The thickness of the graphite coating varied from 4 to 20 μm. The results of this modeling are shown in Fig. 5. The modeling performed showed that the largest deviation of the calculated thermal conductivity coefficient from the true one was observed at the smallest sample thickness (10 μm). In this case, the thickness of the graphite coating h_1 does not affect the obtained values. For such thickness the effect of the pulse finitude becomes more significant than of the graphite coating thickness, since the time of measuring the temperature response on the underside of the sample becomes comparable with the laser pulse width [6,11].

To increase the sample heating time, it is possible to increase the effective thickness of the sample by shifting the detector to a certain distance from the heating center, as suggested in the papers [12,13]. The graphite coating begins to play an important role for samples with thickness of 20 μm or more.

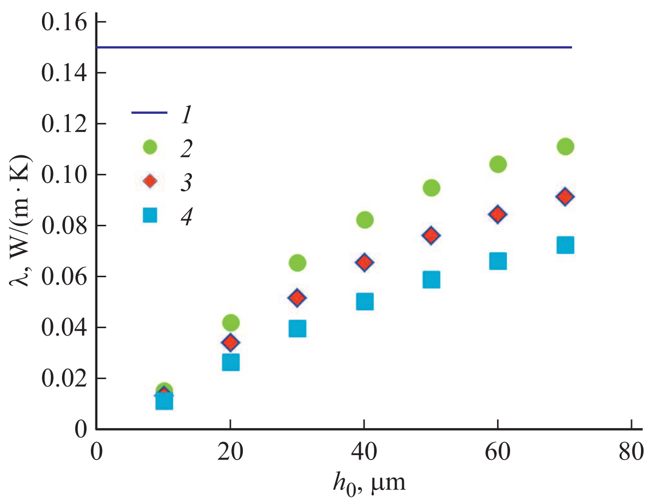

Figure 5. Thermal conductivity coefficient of thin films vs. sample thickness h_0. *1* is true value of λ_t. Graphite coating thickness h_1, μm: *2* — 4, *3* — 10, *4* — 20.

It can also be seen from the graph that the most accurate measurements of thermal conductivity were obtained with the smallest graphite layer, 4 μm, and the largest sample thickness, which is consistent with previous results. For experimental study it is necessary to look for methods of thinner deposition of the graphite coating.

4. Conclusion

The analysis of the modeling results shown that the graphite coating up to 20 μm thick has little effect on the experimental results for the bulk sample. However, repeated coating of the sample with graphite layer with its thickness increasing up to 100 μm leads to the measurements deviation increasing up to 20%.

The thickness of bulk samples does not affect the measurement results. The deviation becomes noticeable when thickness decreases below 0.7 mm. For bulk samples with low thermal conductivity coefficient (< 1 W/(m · K)) the deviation of the value of the thermal conductivity coefficient λ measured by the laser flash method can reach 60%. This can be explained by the presence of a thin layer of graphite on the side surface of the sample, which plays the role of a thermal bridge leading to increased value of the thermal conductivity coefficient. For materials with thermal conductivity coefficient higher than that of graphite, the increasing of the normalized deviation is also observed, but with a different sign, indicating that the presence of the graphite coating leads to the underestimation of measured thermal conductivity coefficient.

For thin samples the contribution of the graphite coating can be much higher (up to 90%). Also, for thin samples the significant parameter that affects the measurement results will be its thickness. When the sample thickness is below 10 μm, the graphite coating thickness will not significantly affect the measurement results. The determining factor will be the power and width of the laser pulse of radiant energy applied to the sample.

Problems arising when measuring thin samples by the laser flash method are related to the heating curve distortion by the laser pulse due to the superposition of the heating temperature curve and the pulse itself. To increase the sample heating time, it is possible to increase the effective thickness of the sample by shifting the detector to a certain distance from the heating center.

When measuring thin samples, it is necessary, if possible, to refuse from additional coatings or to monitor the thickness of the graphite coating for its further consideration in measurements.

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Conflict of interest

The authors declare that they have no conflict of interest.

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