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INVESTIGATION OF THE EFFECT OF RADIATION ON THE PRECISION OF THERMAL CONDUCTIVITY MEASUREMENT BY THE ABSOLUTE METHOD

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- *In this paper, we present the results of computer simulation of a radiation heat exchange between the structural members of measuring setup and the sample when determining the thermal conductivity by the absolute method. Errors due to radiation are determined depending on the emissvity factors of the sample, radiation screen, sample heaters, screen and thermostat, as well as the distance between the screen and sample. Conditions for minimization of radiation effect on the preciison of thermal conductivity measurement are determined.*

Introduction

Precise determination of thermoelectric material properties is important for the development of high-performance thermoelectric devices. Moreover, for quality enhancement of materials proper, there must be a clear relation between the processing steps in the manufacture of materials and the resulting parameters. The existing methods of determining the main thermoelectric properties of materials (thermal conductivity, electric conductivity and thermoEMF) lack precision. The total error of determining the figure of merit by known methods can exceed 10 %, which makes it impossible to solve material science problems, as long as the results of influence on the substance can prove to be smaller than the measurement error.

With regard to possible values of errors, the most complicated process is thermal conductivity measurement over a wide temperature range. It is customary to determine the thermal conductivity of regular-shaped samples using the absolute method [1 – 3].

The main sources of errors in measuring the thermal conductivity by the absolute method are considered in works [4, 5]. They include heat losses from the surface of sample and sample heater, the errors of measuring instruments, the errors of measuring temperature difference on the sample, etc.

The effect of radiation heat losses is considered in works [6, 7]. Heat losses from the sample surface become evident already at a temperature of 200 K, and they should be taken into account when determining the thermal conductivity. To eliminate radiation heat transfer, the sample and sample heater are surrounded by a protective screen with a heater, the power of which is selected such that the temperatures of the sample and screen heaters are identical. Accordingly, temperature distributions along the sample and screen will be approximately identical, too. The volume between the protective screen and the sample is filled with a low thermal conductivity powder. However, in this case corrections should be introduced for heat flux along the isolation powder parallel to the sample.

Another way to reduce the errors due to radiation effect is to take into account radiation heat flux as a correction for total heat flux which is determined as the sample heater power. As long as the value and temperature dependence of sample emissivity factor is not known precisely, in work [7] it is proposed to apply matt blackening on the lateral surface of the sample and to bring closer the emissivity factor of the sample to that of the absolute black body. Moreover, account must be taken of the emissivity factor of measuring setup members. Therefore, the sample was surrounded by a silver polished screen, and heat flux due to heat transfer in the gap between the sample and screen was calculated on condition of absolutely refracting surface of the screen and the lateral surface of the sample similar in properties to the absolute black body.

For a precise account of radiation effect or its minimization it is necessary to establish a relation between the properties of measuring setup members and the sample and possible measurement errors.

The purpose of this research is to determine conditions for minimization of radiation effect on the precision of thermal conductivity measurement by the absolute method.

1. Physical, mathematical and computer models

Sample under study of length l_1 and diameter d_1 is arranged between the sample heater and thermostat, as illustrated in Fig. 1. Arranged around the sample is a radiation screen with a screen heater, the temperatures on the sample and screen ends being identical.

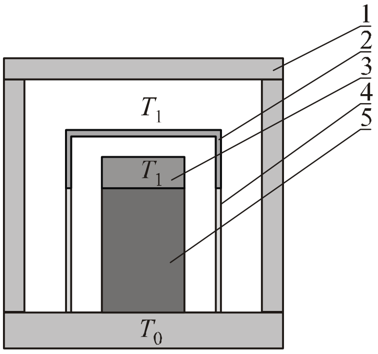

Fig. 1. Schematic of the absolute method of thermal conductivity measurement.
1 – thermostat, 2 – screen heater, 3 – sample heater, 4 – screen, 5 – sample.

Thermostat temperature is T_0, sample and screen heater temperature is T_1. Material thermal conductivity: κ_1 – sample, κ_2 – sample heater, κ_3 – screen, κ_4 – screen heater. Emissivity factors: ε_1 – sample, ε_2 – sample heater, ε_3 – screen, ε_4 – screen heater, ε_5 – thermostat.

Account is taken of heat exchange between the sample, screen, sample heaters and radiation screen, heat transfer due to thermal conductivity along the sample and screen, heat transfer between the radiation screen and thermostat.

To find the temperature distribution, it is necessary to solve the equation of thermal conductivity

$$\nabla(-\kappa \cdot \nabla T) = Q\,, \tag{1}$$

where κ is thermal conductivity, ∇T is temperature gradient, Q is heat flux value.

Limiting conditions taking into account radiation heat exchange between structural members of measuring setup:

- lateral surface of sample

$$r = \frac{d}{2},\ z \in [0;\ l]:\ q = \varepsilon_1 (G_1 - \sigma T^4)\,; \tag{2}$$

- lateral surface of sample heater

$$r = \frac{d}{2},\ z \in [l;\ l + l_2]:\ q = \varepsilon_2 (G_2 - \sigma T^4)\,; \tag{3}$$

- upper surface of sample heater

$$r \in \left[0;\ \frac{d}{2}\right],\ z = l + l_2:\ q = \varepsilon_2 (G_3 - \sigma T^4)\,; \tag{4}$$

- internal surface of screen

$$r=\frac{d}{2}+\Delta R,\ z\in[0;\ l]:\ q=\varepsilon_3(G_4-\sigma T^4);\tag{5}$$

- internal surfaces of screen heater

$$r=\frac{d}{2}+\Delta R,\ z\in[l;\ l+l_2]:\ q=\varepsilon_4(G_5-\sigma T^4);\tag{6}$$

$$r\in\left[0;\ \frac{d}{2}+\Delta R\right],\ z=l+l_2+\Delta R:\ q=\varepsilon_4(G_6-\sigma T^4);\tag{7}$$

- external surfaces of screen heater

$$r=\frac{d_4}{2},\ z\in[l;\ l+l_4]:\ q=\varepsilon_4(G_7-\sigma T^4);\tag{8}$$

$$r\in\left[0;\ \frac{d}{2}+\Delta R\right],\ z=l+l_4:\ q=\varepsilon_4(G_8-\sigma T^4),\ T=T_0+\Delta T;\tag{9}$$

- external surface of screen

$$r=\frac{d_3}{2},\ z\in[0;\ l]:\ q=\varepsilon_3(G_9-\sigma T^4);\tag{10}$$

- thermostat surface between the sample and screen

$$r\in\left[\frac{d}{2};\ \frac{d}{2}+\Delta R\right],\ z=0:\ q=\varepsilon_5(G_{10}-\sigma T^4);\tag{11}$$

- thermostat surfaces on the outside of the screen

$$r\in\left[\frac{d_3}{2};\ \frac{d_5}{2}\right],\ z=0:\ q=\varepsilon_5(G_{11}-\sigma T^4);\tag{12}$$

$$r\in\left[0;\ \frac{d_5}{2}\right],\ z=l_5:\ q=\varepsilon_5(G_{12}-\sigma T^4);\tag{13}$$

$$r=\frac{d_5}{2},\ z\in[0;\ l_5]:\ q=\varepsilon_5(G_{13}-\sigma T^4);\tag{14}$$

- external surface of thermostat

$$r\in\left[0;\ \frac{d_5}{2}+h_5\right],\ z=-h_5:\ T=T_0;\tag{15}$$

$$r\in\left[0;\ \frac{d_5}{2}+h_5\right],\ z=l_5+h_5:\ T=T_0;\tag{16}$$

$$r=\frac{d_5}{2}+h_5,\ z\in[-h_5;\ l_5+h_5]:\ T=T_0.\tag{17}$$

where d_1, l_1 is diameter and length of sample, d_2, l_2 is diameter and length of sample heater, d_3, l_3 is outside diameter and length of screen, d_4, l_4 is diameter and length of screen heater, d_5, l_5, h_5 is internal diameter, length and thickness of thermostat, ΔR is the distance between the sample and screen, ε_{1-5} are emissivity factors of surfaces, σ is Stephan-Boltzmann constant, G is incoming radiation heat flux for each separate boundary:

$$G=G_m+F_{amb}\sigma T_{amb}^4,\tag{18}$$

where G_m is radiation value from the other boundaries of measuring setup and sample, F_{amb} is ambient view factor, T_{amb} is temperature at a distant point in directions included into F_{amb}.

Finding a solution to equation (1) with the boundary conditions (2) – (17) is an extremely complicated problem, and even if it can be solved analytically, the resulting solutions will be too complicated and not subject to further analysis. Therefore, to find the distributions of temperature and heat fluxes, a package of applied programs COMSOL Multiphysics was employed in the measuring system. With regard to problem symmetry, calculations were made for a 2 *d*-model with axial symmetry. The resulting solutions of thermal conductivity equation in the sample and structural members of measuring setup do not possess the universality of analytical solutions, but still they permit solving concrete optimization problems for measuring setups intended for determining the thermal conductivity of samples of given geometrical shape and dimensions in the assigned temperature range.

Coefficient G_m which depends on the mutual arrangement of surfaces, is calculated by introducing into computer model of additional variable J which is assigned by equation

$$J = (1-\varepsilon)\left\{G_m(J) + F_{amb}\sigma T_{amb}^4\right\} + \varepsilon\sigma T^4, \tag{19}$$

solved together with thermal conductivity equation.

2. The results of computer simulation of temperature and heat flux distributions

Fig. 2 shows typical distributions of temperature and heat fluxes in the sample and structural members of the measuring setup.

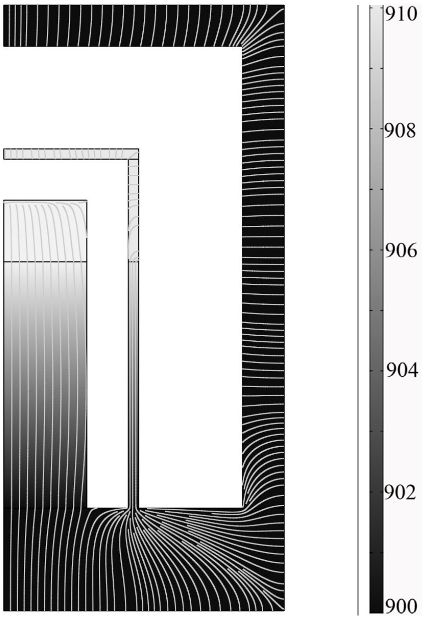

Fig. 2. Temperature and heat flux distribution due to thermal conductivity in the sample and structural members of the measuring setup.

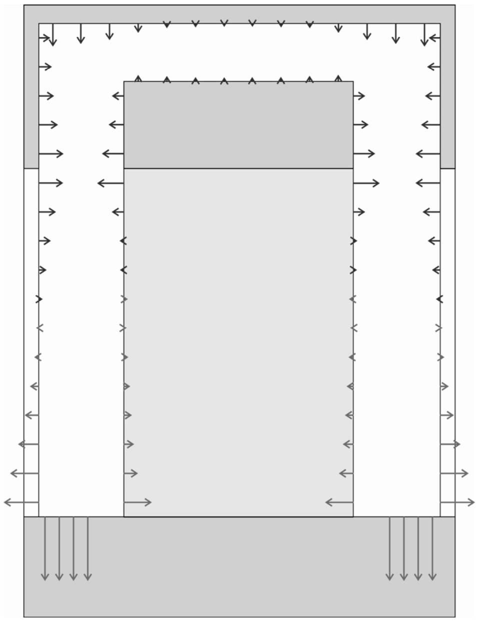

Fig. 3. Heat flux distribution due to radiation in the sample, screen and sample and screen heaters.

The distribution of radiation heat fluxes from the surface of sample, screen, sample heater and screen heater is shown in Fig. 3. The arrow size characterizes the value of heat flux density and the arrow direction designates the absorption or radiation of heat.

As can be seen from Fig. 3, even with the identical temperatures of the sample and screen heaters, as well as the identical temperature distributions along the sample and screen, there is heat transfer from the upper parts of the sample and screen, as well as the sample and screen heaters, to thermostat and lower parts of the sample and screen. The values of these fluxes are shown in Figs. 4, 5 (for typical sample length 12 mm and diameter 8 mm with thermal conductivity 2 W/(m·K) and reliable emissivity factors of sample $\varepsilon_1 = 0.8$, heaters, thermostat and screen $\varepsilon_2 = \varepsilon_3 = \varepsilon_4 = \varepsilon_5 = 0.5$).

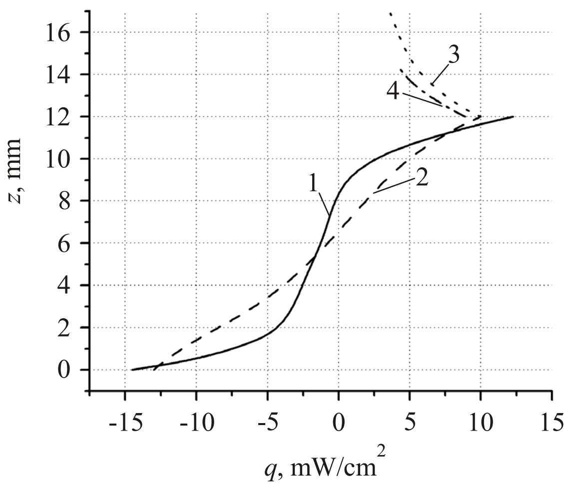

Fig. 4. Densities of radiation heat flux from the surface of sample, screen and the lateral surface of heaters. 1 – radiation from sample surface; 2 – radiation from screen surface; 3 – radiation from screen heater surface; 4 – radiation from sample heater surface.

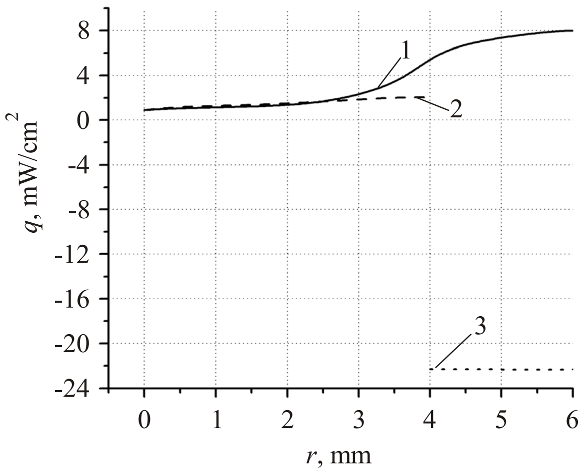

Fig. 5. Densities of radiation heat flux from the surface of heaters and thermostat. 1 – radiation from screen heater surface; 2 – radiation from sample heater surface; 3 – radiation from thermostat surface.

Temperature distribution along the sample and screen and their deviation from the linear distribution is shown in Fig. 6.

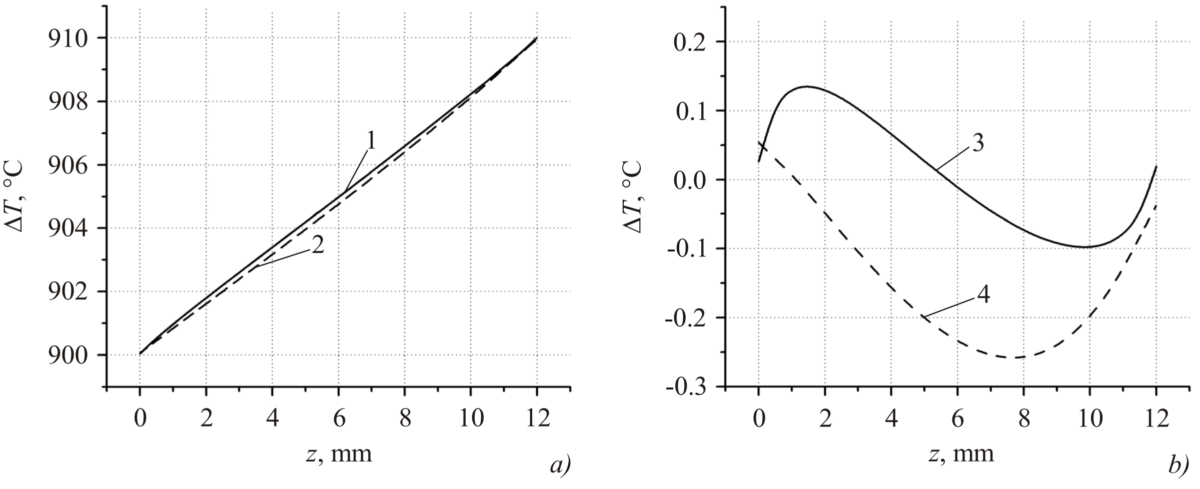

Fig. 6. Temperature distribution along the sample and screen (a) and their deviation from the linear distribution (b). 1, 3 – sample, 2, 4 – screen.

3. The results of the investigation of radiation effect on the precision of thermal conductivity measurement and their analysis

The elaborated computer model was used to obtain distributions in the sample and the structural members of the measuring setup intended for thermal conductivity measurement in the temperature range of 300 to 900 K for the samples of typical length 12 mm and diameter 8 mm. Dependence of the thermal conductivity errors on the emissivity factors of the sample, screen, sample and screen heaters, thermostat, as well as the distance between the sample and screen and sample thermal conductivity has been investigated. The results of the investigation are given below. If necessary, computer model allows reproducing these results for alternative temperature ranges and sample dimensions.

3.1. Effect of screen and screen heater emissivity factors

Figs. 7, 8 show dependences of the thermal conductivity errors on the emissivity factors of the screen and screen heater for the following parameter values of the sample and measuring setup structural members: sample thermal conductivity 2 W/(m·K), sample length 12 mm, diameter 8 mm, the distance between the sample and screen 2 mm, the emissivity factors $\varepsilon_1 = 0.8$; $\varepsilon_2 = 0.5$; $\varepsilon_5 = 0.5$.

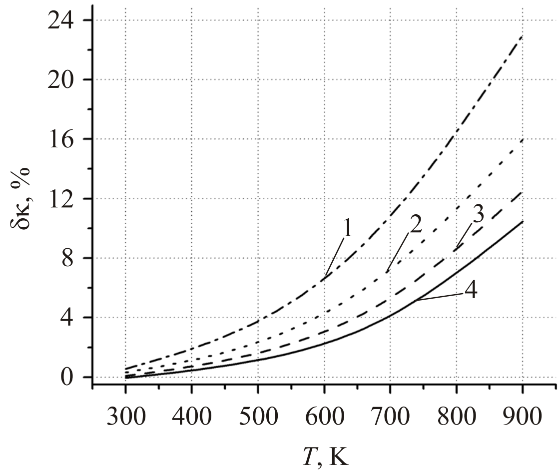

Fig. 7. Temperature dependences of the thermal conductivity errors for different emissivity factors of the screen and screen heater ($\varepsilon_3 = \varepsilon_4$: 1 – 0.1, 2 – 0.3, 3 – 0.5, 4 – 0.7).

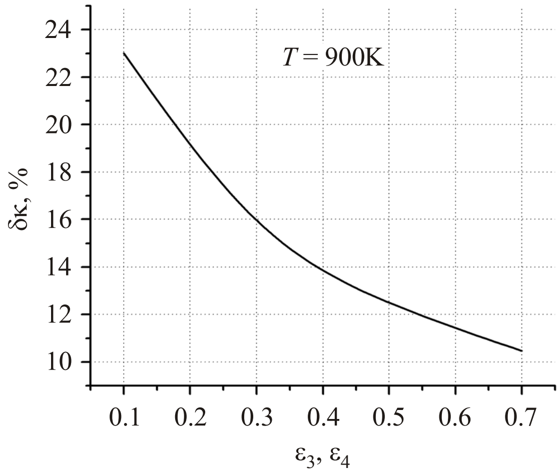

Fig. 8. Dependence of the thermal conductivity error on the emissivity factors of the screen and screen heater

As can be seen from Figs. 7, 8, for larger values of screen and screen heater emissivity factors the thermal conductivity errors will be smaller. Therefore, to reduce these errors, the surfaces of the screen and screen heater should be possibly selected with as large emissivity factors as possible.

3.2. Effect of thermostat emissivity factor

Figs. 9, 10 show dependences of the thermal conductivity errors on thermostat emissivity factor for the following parameter values of the sample and measuring setup structural members: sample thermal conductivity 2 W/(m·K), sample length 12 mm, diameter 8 mm, the distance between the sample and screen 2 mm, the emissivity factors $\varepsilon_1 = 0.1$; $\varepsilon_2 = 0.1$; $\varepsilon_3 = 0.5$; $\varepsilon_4 = 0.5$.

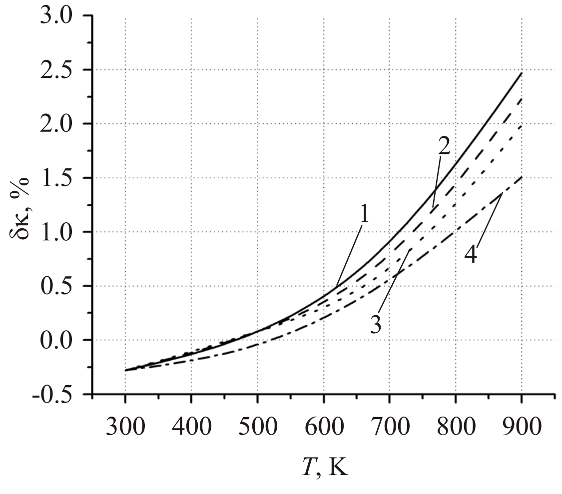

Fig. 9. Temperature dependences of the thermal conductivity errors for different values of thermostat emissivity factor. (ε_5: 1 – 0.7, 2 – 0.5, 3 – 0.3, 4 – 0.1)

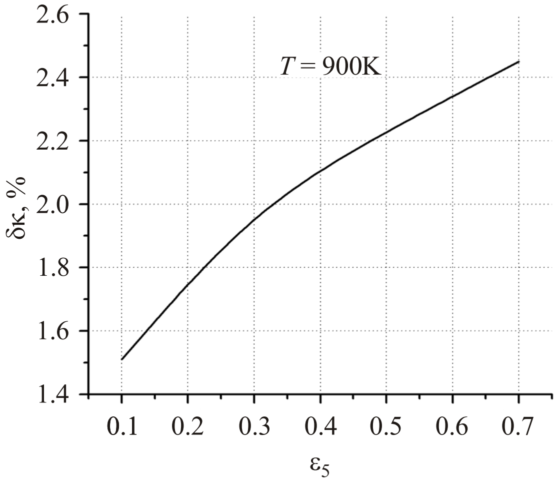

Fig. 10. Dependence of the thermal conductivity error on thermostat emissivity factor.

Thus, the surface properties of thermostat, unlike the properties of the screen and screen heater, have an opposite effect on the accuracy of thermal conductivity determination, namely for higher values of thermostat emissivity factor the errors will be larger.

3.3. Effect of sample and sample heater emissivity factors

Dependences of the thermal conductivity errors for different sample emissivity factors are shown in Figs. 11, 12. Sample thermal conductivity is 2 W/(m·K), sample length is 12 mm, diameter is 8 mm, the distance between sample and screen is 2 mm, the emissivity factors are: $\varepsilon_2 = 0.1$; $\varepsilon_3 = 0.5$; $\varepsilon_4 = 0.5$ $\varepsilon_5 = 0.5$.

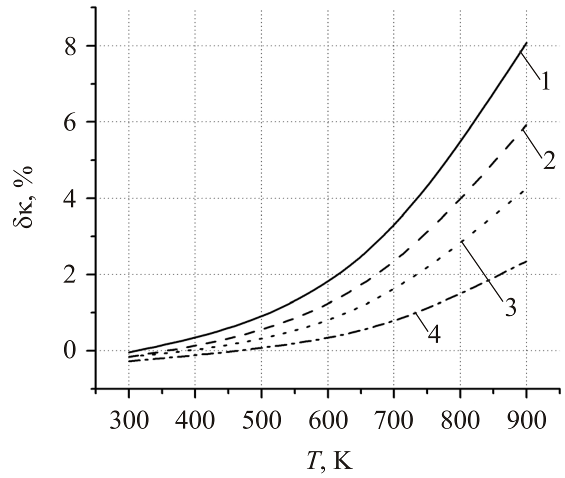

Fig. 11. Temperature dependences of the thermal conductivity errors for different sample emissivity factors. (ε_1: 1 – 0.8, 2 – 0.5, 3 – 0.3, 4 – 0.1).

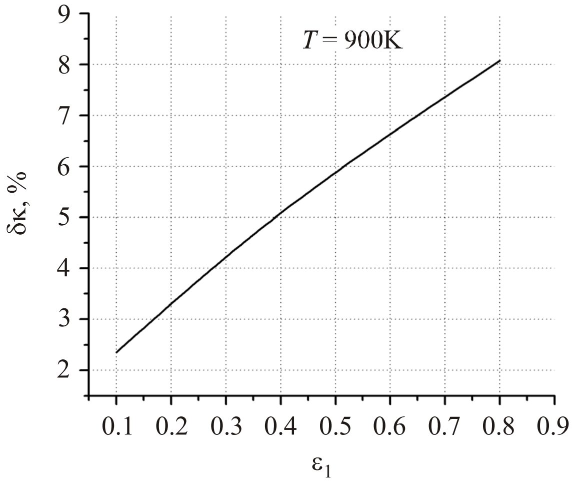

Fig. 12. Dependence of the thermal conductivity error on the sample emissivity factor.

Dependences of the thermal conductivity errors on the emissivity factor of sample heater for different emissivity factors of screen, screen heater and thermostat is shown in Fig. 13. The sample has thermal conductivity 2 W/(m·K), length 2 mm, diameter 8 mm, the distance between the sample and screen is 2 mm, the sample emissivity factor $\varepsilon_1 = 0.8$.

Thus, according to the results of computer studies for sample and sample heater surfaces with large emissivity factors, the thermal conductivity errors related to radiation heat exchange will have large values. Therefore, one should choose possibly lower emissivity factors of the sample and sample heater, just as thermostat emissivity factor.

3.4. Effect of sample thermal conductivity

Fig. 14 shows a change in measurement error with a change in thermal conductivity of sample being studied, if the emissivity factors of sample and structural members of measuring setup are known ($\varepsilon_1 = 0.8$; $\varepsilon_2 = 0.5$; $\varepsilon_3 = 0.5$; $\varepsilon_4 = 0.5$; $\varepsilon_5 = 0.5$).

Thus, the effect of radiation on the measured results is most pronounced for bad heat conductors. With increasing sample thermal conductivity, this effect is minimized.

3.5. Effect of the distance between the sample and screen

To determine the effect of the distance between the sample and screen on the accuracy of thermal conductivity measurement, we investigated measurement errors and obtained their temperature dependences for different emissivity factors of the sample, sample heater, screen, screen heater and thermostat with the distances between the sample and screen from 1 mm to 2 mm.

Dependences of the thermal conductivity errors on the distance Δ*r* between the sample and screen for different thermostat temperature values are shown in Fig. 15, 16. The sample has thermal conductivity 2 W/(m·K), diameter 8 mm, length 12 mm. As is evident from the plots, these dependences are linear.

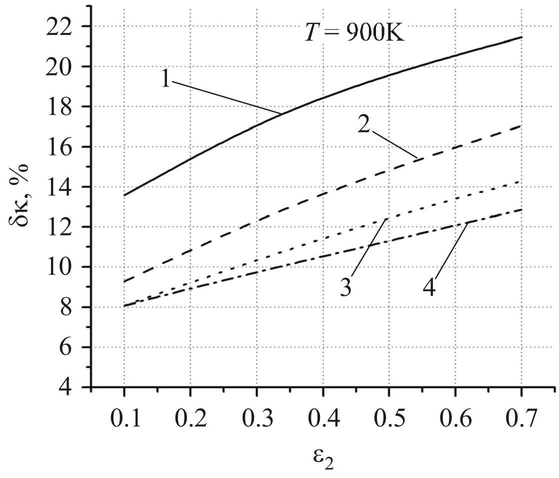

Fig. 13. Dependences of the thermal conductivity error on the emissivity factor of sample heater for different emissivity factors of screen, screen heater and thermostat. ($\varepsilon_3 = \varepsilon_4 = \varepsilon_5$: *1 – 0.1, 2 – 0.3, 3 – 0.5, 4 – 0.7).*

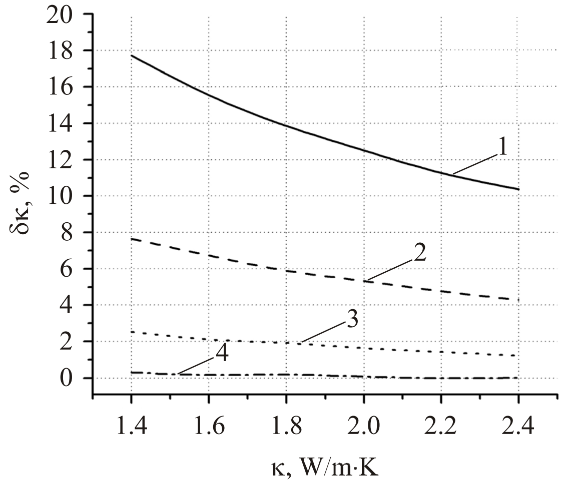

Fig. 14. Dependences of the thermal conductivity error on the sample thermal conductivity for different thermostat temperature values (1 – T = 900 K; 2 – T = 700 K; 3 – T = 500 K; 4 – T = 300 K).

As can be seen from Fig. 15 – 16, dependences of the thermal conductivity errors on the distance between the sample and screen are linear for different operating temperatures and properties of sample surfaces and structural members of measuring setup. We also emphasize that with increasing distance between the sample and screen, the thermal conductivity errors grow linearly, so the screen must be arranged as close to the sample as the design of measuring setup permits.

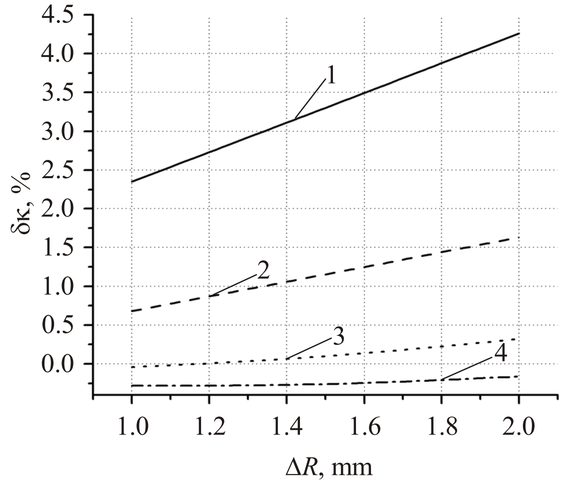

Fig. 15. Dependences of the thermal conductivity errors on the distance between the sample and screen Δ*r for different thermostat temperature values (emissivity factors:* $\varepsilon_1 = 0.3$; $\varepsilon_2 = 0.1$; $\varepsilon_3 = 0.5$; $\varepsilon_4 = 0.5$; $\varepsilon_5 = 0.5$).

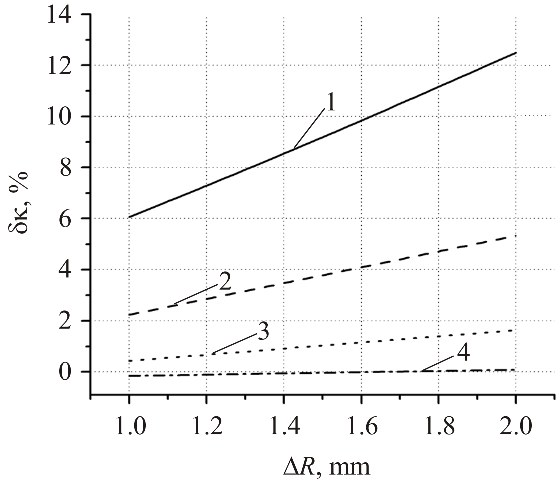

Fig. 16. Dependences of the thermal conductivity errors on the distance between the sample and screen Δ*r for different thermostat temperature values (emissivity factors:* $\varepsilon_1 = 0.8$; $\varepsilon_2 = 0.5$; $\varepsilon_3 = 0.5$; $\varepsilon_4 = 0.5$ $\varepsilon_5 = 0.5$).

1 – T = 900 K; 2 – T = 700 K; 3 – T = 500 K; 4 – T = 300 K.

Based on such results, for instance, for measuring the thermal conductivity of *Bi*-*Te*-based materials, we used the emissivity factors: $\varepsilon_1 = 0.1$; $\varepsilon_2 = 0.1$; $\varepsilon_3 = 0.9$; $\varepsilon_4 = 0.9$; $\varepsilon_5 = 0.1$ and the distance between the screen and sample $\Delta R = 1$ mm. The thermal conductivity error related to radiation heat exchange for such values does not exceed 0.4 % in the temperature range of 300 – 800 K.

Conclusions

1. It was determined that even in the case when the temperatures of the sample and screen heaters, as well as the temperature distributions along the sample and screen are identical, there is essential heat transfer from the upper parts of the sample, screen and sample and screen heaters to the lower parts of the sample, screen and thermostat. In the absence of special measures aimed at reducing such heat transfer, the thermal conductivity errors can reach 15 %.
2. Computer model of radiation heat transfer was elaborated to determine the thermal conductivity errors with a radiation screen.
3. Temperature dependences of the thermal conductivity errors on the emissivity factors of the sample, sample heater, screen, screen heater and thermostat were obtained. For typical dimensions of measuring samples it was determined that the most considerable contribution to the error is made by the sample and sample heater emissivity factors. In so doing, the emissivity factor value of the sample and sample heater must be selected possibly lower, and that of the screen and screen heater – possibly higher.
4. Temperature dependences of the thermal conductivity errors for different distances between the sample and screen were obtained. It was determined that such dependence is linear, and the thermal conductivity error is reduced with decreasing the distance between the screen and sample.
5. Conditions were determined that allow minimizing the effect of radiation on the accuracy of determining the thermal conductivity when measuring *Bi*-*Te* material samples. The thermal conductivity error due to radiation heat exchange, in case when the recommended measures are taken, does not exceed 0.4 % .

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