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METHOD FOR MEASURING THE INTEGRAL RADIATION, TRANSMISSION, AND REFLECTION COEFFICIENTS FOR SELECTIVE TRANSLUSCENT MATERIALS

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UDC 535.3

An account is given of the main conclusions and results of testing a method for the determination of the integral radiative properties of materials.

The development of many segments of the national economy (machine building, power industry, construction industry, etc.) requires the use of new materials not only with desired mechanical, electrical, and thermal properties but also with known optical properties. It is impossible to obtain such materials without controlling optical production parameters both at the production stage and during operation as parts of products or structures. In turn, a highly efficient control and measurement instrument cannot be created without refining measurement methods.

The (spectrally) integrated radiation, transmission, and reflection coefficients of materials are among the most important opticophysical quantities characterizing the material properties that are to be measured or controlled. The goal of the present report is to validate a new method of measurement of the named integral coefficients which is applicable to spectrally selective transluscent materials at moderate, close to normal, temperatures.

The optical methods and instruments for the measurement of radiation (or absorption), transmission, and reflection coefficients of transluscent light-diffusing materials at high [1, 2], medium, and reduced [3-5] temperatures are known. Thus, according to [4], a plane sample of a material thermally stabilized at temperature T_2 is placed in an isothermal chamber which simulates a blackbody cavity at a temperature $T_1 \neq T_2$ and, using a photodetection system (PDS), the irradiances of chamber wall L_1 and of sample L_{11} on top of the chamber background are measured. Next, a blackbody at temperature T_2 is placed behind the sample and the irradiance of sample L_{12} is measured on top of the blackbody background. After removing the sample the irradiance of the blackbody L_2 is measured. These results determine the quantities

$$\varepsilon = (L_{11} - L_1)/(L_2 - L_1), \tag{1}$$

$$\varepsilon + \tau = (L_{12} - L_1)/(L_2 - L_1). \tag{2}$$

Method [4] has a limitation related to its applicability for only carrying out spectral measurements and to the impossibility (or to large errors) of measurements of the integral quantities ε, τ, and ρ for spectrally selective materials. The latter is a consequence of the temperature dependence of these integral quantities. Thus, due to the different relative spectral irradiance blackbody distributions at temperatures T_1 and T_2, $L(\lambda, T_1)/T_1^4 \neq L(\lambda, T_2)/T_2^4$ even if the spectral coefficients are temperature independent:

$$\begin{aligned} \varepsilon(\lambda, T_1) &= \varepsilon(\lambda, T_2) = \varepsilon(\lambda), \\ \tau(\lambda, T_1) &= \tau(\lambda, T_2) = \tau(\lambda), \\ \rho(\lambda, T_1) &= \rho(\lambda, T_2) = \rho(\lambda) \end{aligned} \tag{3}$$

so that the following inequalities hold true:

$$\varepsilon(T_1) \neq \varepsilon(T_2), \quad \tau(T_1) \neq \tau(T_2), \quad \rho(T_1) \neq \rho(T_2). \tag{4}$$

Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 62, No. 3, pp. 446-452, March, 1992. Original article submitted May 12, 1991.

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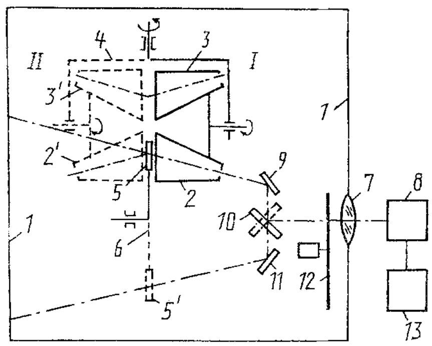

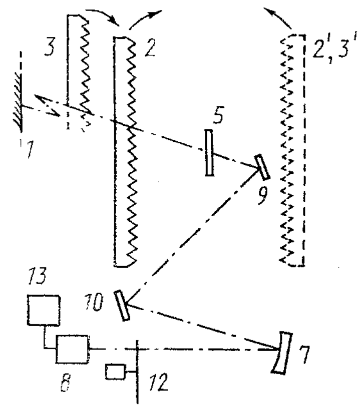

Fig. 1 Fig. 2

Fig. 1. Schematic optical diagram of the device for implementing the measurement method.

Fig. 2. Optical diagram of the apparatus used for testing the measurement method. The designations are the same as in Fig. 1.

In order to illustrate the consequences of not taking into account the temperature dependence of $\varepsilon(T)$ during integral measurements by the method of [4], express the irradiance, L_{11}, using the relation

$$\varepsilon(T_1) + \tau(T_1) + \rho(T_1) = 1, \tag{5}$$

which follows from the energy conservation law, Kirchhoff's law, and – for metals [1-5] – also from the Helmholtz invertibility principle in the form $L_{11} = \varepsilon(T_2)L_2 + \tau(T_1)L_1 + \rho(T_1)\cdot L_1 = \varepsilon(T_2)L_2 + [1 - \varepsilon(T_1)]L_1$. Substituting this expression into Eq. (1), we obtain

$$\varepsilon = \frac{\varepsilon(T_2)L_2 - \varepsilon(T_1)L_1}{L_2 - L_1}. \tag{6}$$

It follows from Eq. (6) that for $\varepsilon(T)$ which depends monotonically on temperature the calculated quantities ε are not within the interval between $\varepsilon(T_1)$ and $\varepsilon(T_2)$ except in the case when $\varepsilon(T_1) = \varepsilon(T_2) = \varepsilon$. Since the latter case corresponds only to nonselective (gray) materials ($\varepsilon(\lambda) = \text{const}$), the errors in applying the method [4] to the measurement of ε, τ, and ρ for spectrally selective materials become evident.

The methodology of [5] for measuring the integral quantities ε, τ, and ρ for materials leads to similar errors since it also employs blackbodies at (three) different temperatures with a subsequent change of conditions under which these blackbodies illuminate both sides of the sample. According to this method, the spectral coefficients ε, ρ, and τ are defined by the expressions

$$\varepsilon = 1 - A, \quad \tau = (A + B)/2, \quad \rho = (A - B)/2, \tag{7}$$

where

$$A = \frac{L_{32} + L_{23} - 2L_{11}}{L_2 + L_3 - 2L_1}, \quad B = \frac{L_{32} - L_{23}}{L_2 - L_3}. \tag{8}$$

Thus, the reviewed and other known measurement methods do not guarantee that the goal that was posed earlier will be accomplished. The solution to the problem can be achieved by the method presented below. As will be shown, the solution is physically not exact but is obtained with two assumptions which insignificantly limit the applicability of the method.

The presentation of the essence of the method, which is the basis of the method of [5], will be conducted using the optical instrument diagram shown in Fig. 1. In isometric chamber 1, whose cavity simulates a blackbody with a given temperature T_1, two blackbodies – 2 and 3 – with temperatures T_2 and T_3 which are not equal to T_1 are placed. These blackbodies are attached to revolving unit 4 which, by alternately turning around two axes, produces an interchange in the positions of blackbodies 2 and 3 as well as in their rotation by 180°. Blackbodies 2 and 3 are placed with radiative apertures to one side. Measured sample 5 is placed at a minimum distance in front of one of the blackbodies. Being attached to moveable device 6, the sample can be moved away from the blackbodies (position 5') so that it is irradiated only by walls of chamber 1. Just as in [5], blackbodies 2 and 3 have one

TABLE 1. Results of Measurements during Method Testing

Measured and calculated values	Variants		
	1	2	3
T_1, °C(K)	21(294)	21(294)	21(294)
T_2, °C(K)	50(323)	50(323)	65(338)
T_3, °C(K)	80(353)	65(338)	80(353)
U_1, V	0	0	0
U_2, V	0,717	0,717	1,172
U_3, V	1,692	1,172	1,692
U_{11}, V	0	0	0
U_{12}, V	0,332	0,332	0,533
U_{13}, V	0,758	0,533	0,758
U_{21}, V	0,089	0,089	0,144
U_{31}, V	0,207	0,144	0,207
$\tau(T_1)$	0,529	0,537	0,519
$\rho(T_1)$	0,132	0,135	0,128
$\varepsilon(T_1)$	0,339	0,328	0,353
t, 10^{-4} K^{-1}	—7,5	—8,5	—6,5
r, 10^{-4} K^{-1}	—0,9	—1,3	—0,5
e, 10^{-4} K^{-1}	8,4	9,8	7,0
τ(80°C)	0,485	0,487	0,481
ρ(80°C)	0,126	0,127	0,125
ε(80°C)	0,389	0,386	0,394

through-hole on their radiating surfaces for sighting the side of the sample that is irradiated by the given blackbody. These openings in the operating positions of each of the blackbodies are located on the axis of the PDS, which consists of objective lens 7, spectrally nonselective radiation detector 8 which transforms the radiation it absorbs into electrical signals, stationary plane mirror 9, and reversible plane mirror 10 which also can direct the field of vision of the PDS through second fixed plane mirror 11 onto the sample in position 5' (or, in the absence of the sample, onto chamber wall 1). In order to exclude the interference effect of blackbodies 2 and 3 on the irradiance of sample 5 and of chamber walls 1, screens (not shown on the diagram) at temperature T_1 are placed inside the chamber. Chopper 12 and recording instrument 13 are part of the PDS.

Measurements are performed as follows. Temperatures T_1, T_2, and T_3 of all three blackbodies are established, stabilized, and measured. It is desirable that these temperatures differ from one another by no less than a few tens of degrees, which is necessary in order to create and detect a reliably reproducible signal level at the output of the PDS. Sample 5 is fixed in holder 6 and is placed in position 5'; at the same time, sample temperature T_0 assumes a value close (almost equal) to chamber temperature T_1. Mirror 10 is fixed in the position denoted by dashed lines, and the irradiance, L_{11}, of sample 5' is measured under the conditions of isotropic irradiation from both sides by chamber walls 1 – that is, by blackbodies at the first (assigned) temperature T_1. Next, sample 5' is taken away from the field of view of the PDS and the irradiance, L_1, of a blackbody at temperature T_1 is measured.

Blackbodies 2 and 3 are fixed in position 1 (to the right of the sample, on its visible side), mirror 10 is moved to the position given by the solid lines, sample 5 is placed in front of the aperture of blackbody 2, and, through its opening, the irradiance, L_{21}, of the sample is measured. Next, using revolving device 4, blackbodies 2 and 3 switch places and the irradiance, L_{31}, of the sample is measured. After that, using the same device, 4, the blackbodies are placed in position II (on the diagram it is to the left of the sample, on its reverse side). After the placement of blackbody 3 directly behind the sample, the irradiance, L_{13}, of the sample is measured, and, after switching the positions of blackbodies 2 and 3, the irradiance, L_{12}, of the sample is measured. Next, sample 5 is removed from the PDS field of view and the irradiance, L_2, of blackbody 2 is measured; blackbodies 2 and 3 switch places again and the irradiance, L_3, of blackbody 3 is measured.

Based on the linear transformation of radiation during transmission and reflection and on the absence of multiple reflections between the blackbodies and the sample and also taking into account that, in general case $T_0 \neq T_1$, we write the functional relations between the measured integral irradiance and the sought integral coefficients as

$$L_{11} = \varepsilon(T_0) L_0 + \tau(T_1) L_1 + \rho(T_1) L_1, \quad (9)$$

$$L_{12} = \varepsilon(T_0) L_0 + \tau(T_2) L_2 + \rho(T_1) L_1, \tag{10}$$

$$L_{13} = \varepsilon(T_0) L_0 + \tau(T_3) L_3 + \rho(T_1) L_1, \tag{11}$$

$$L_{21} = \varepsilon(T_0) L_0 + \tau(T_1) L_1 + \rho(T_2) L_2, \tag{12}$$

$$L_{31} = \varepsilon(T_0) L_0 + \tau(T_1) L_1 + \rho(T_3) L_3. \tag{13}$$

In addition to these relations, Eqs. (9)-(13), and the identity, Eq. (5), the sought quantities are related by expressions similar to Eq. (5) at two other temperatures as follows:

$$\varepsilon(T_2) + \tau(T_2) + \rho(T_2) = 1, \tag{14}$$

$$\varepsilon(T_3) + \tau(T_3) + \rho(T_3) = 1. \tag{15}$$

The system of the eight equations, Eqs. (5) and (9)-(15), contain 11 unknown quantities (two of them – $\varepsilon(T_0)$ and L_0 – are indeterminate and nine of them – ε, τ, and ρ at three temperatures each – are unknown) and cannot be solved in a general form. In order to determine the integral coefficients – $\varepsilon(T_1)$, $\tau(T_1)$, and $\rho(T_1)$ – on the basis of this data, we use a property that has been confirmed by experimental and numerical studies and that establishes an almost linear dependence of the sought quantities on temperature. Such a dependence occurs especially in a relatively narrow interval of employed blackbody temperatures (maximally differing from one another by magnitudes on the order of no more than 100 K). In this case we may write

$$\varepsilon(T_2) = \varepsilon(T_1) + e(T_2 - T_1), \quad \varepsilon(T_3) = \varepsilon(T_1) + e(T_3 - T_1), \tag{16}$$

$$\tau(T_2) = \tau(T_1) + t(T_2 - T_1), \quad \tau(T_3) = \tau(T_1) + t(T_3 - T_1), \tag{17}$$

$$\rho(T_2) = \rho(T_1) + r(T_2 - T_1), \quad \rho(T_3) = \rho(T_1) + r(T_3 - T_1). \tag{18}$$

After calculating in pairs the differences L_{12}-L_{11}, L_{13}-L_{11}, and L_{21}-L_{11}, L_{31}-L_{11} for L_0 = const and using the relations in Eqs. (17) and (18), two systems of equations result:

$$\begin{aligned} L_{12} - L_{11} &= \tau(T_2) L_2 - \tau(T_1) L_1 = \tau(T_1)(L_2 - L_1) + tL_2(T_2 - T_1), \\ L_{13} - L_{11} &= \tau(T_3) L_3 - \tau(T_1) L_1 = \tau(T_1)(L_3 - L_1) + tL_3(T_3 - T_1), \end{aligned} \tag{19}$$

$$\begin{aligned} L_{21} - L_{11} &= \rho(T_2) L_2 - \rho(T_1) L_1 = \rho(T_1)(L_2 - L_1) + rL_2(T_2 - T_1), \\ L_{31} - L_{11} &= \rho(T_3) L_3 - \rho(T_1) L_1 = \rho(T_1)(L_3 - L_1) + rL_3(T_3 - T_1), \end{aligned} \tag{20}$$

each of which contains two unknown quantities – $\tau(T_1)$, t (19) and ρ (T_1), r (20). By making a simple transformation of Eqs. (19) and (20) and using the equality e + t + r = 0, which comes from the identity, Eq. (5), and the relations determining the integral irradiance of blackbodies $L_2 = (\sigma/\pi)T_2^4$ and $L_3 = (\sigma/\pi)T_3^4$, we find that

$$\tau(T_1) = \frac{(L_{12} - L_{11}) T_3^4 (T_3 - T_1) - (L_{13} - L_{11}) T_2^4 (T_2 - T_1)}{(L_2 - L_1) T_3^4 (T_3 - T_1) - (L_3 - L_1) T_2^4 (T_2 - T_1)}, \tag{21}$$

$$\rho(T_1) = \frac{(L_{21} - L_{11}) T_3^4 (T_3 - T_1) - (L_{31} - L_{11}) T_2^4 (T_2 - T_1)}{(L_2 - L_1) T_3^4 (T_3 - T_1) - (L_3 - L_1) T_2^4 (T_2 - T_1)}, \tag{22}$$

$$\varepsilon(T_1) = 1 - \tau(T_1) - \rho(T_1), \tag{5'}$$

$$t = \frac{(L_{13} - L_{11})(T_2^4 - T_1^4) - (L_{12} - L_{11})(T_3^4 - T_1^4)}{(L_2 - L_1) T_3^4 (T_3 - T_1) - (L_3 - L_1) T_2^4 (T_2 - T_1)}, \tag{23}$$

$$r = \frac{(L_{31} - L_{11})(T_2^4 - T_1^4) - (L_{21} - L_{11})(T_3^4 - T_1^4)}{(L_2 - L_1) T_3^4 (T_3 - T_1) - (L_3 - L_1) T_2^4 (T_2 - T_1)}, \tag{24}$$

$$e = -(t + r). \tag{25}$$

As shown in [5], given the linearity and constancy of the sensitivity parameters of the PDS (including chopper 12 which is functionally associated with it), the ratios of the differences of the measured irradiance can be replaced by the ratios of the differences of electrical signals corresponding to each of the measured irradiances. For example, the mathematical formula for the determination of $\tau(T_1)$ assumes the form

$$\tau(T_1) = \frac{(U_{12}-U_{11})\,T_3^4\,(T_3-T_1)-(U_{13}-U_{11})\,T_2^4\,(T_2-T_1)}{(U_2-U_1)\,T_3^4\,(T_3-T_1)-(U_3-U_1)\,T_2^4\,(T_2-T_1)}. \tag{21'}$$

Equations (22)-(24) acquire a similar form.

Thus, using Eqs. (21)-(25) together with Eq. (5') and the results of the measurements of output PDS signals corresponding to the irradiance of the sample under the conditions of the present method – i.e., irradiating the sample from two sides with blackbodies, whose irradiances are known, at three known temperatures – the integral radiation, transmission, and reflection coefficients of the sample at one (the first) of the blackbody temperatures as well as the gradients of these coefficients can be determined. Using Eqs. (16)-(18), the latter circumstance allows for the determination of these coefficients at other temperatures in the temperature range of the employed blackbodies.

The proposed method was approved on a prototype apparatus similar to that used in [5], whose diagram is shown in Fig. 2. The temperature of the enclosure which simulates chamber 1 was equal to $T_1 = 21°C$. For radiation detector 8 we used a BSG-2-type bolometer with a KRS-5 crystal window which is transparent between the wavelengths of 0.6-40 μm. With blackbodies 2 and 3 and sample 5 removed, signal U_1, corresponding to the irradiance of a blackbody at temperature T_1, is measured. This signal was equal to the noise at the output of digital recording apparatus 13 (U_1 = 2-3 mV) and is assumed to be zero. Afterwards, blackbodies 2 and 3 are introduced one at a time into the field of view of the PDS and signals U_2 and U_3, corresponding to their irradiances, are measured. Next, again removing blackbodies 2 and 3, sample 5 is placed in the field of view of the PDS, covering it fully, and signal U_{11}, which corresponds to the irradiance of a sample irradiated from both sides by blackbodies at temperature T_1, is measured (this signal is also equal to zero since the sample temperature coincided with the temperature of the enclosure). After this, blackbody 2 is put at a mounting place (in Fig. 2, to the left of the sample for no longer than 5 sec) and signal U_{12} is measured; next, blackbody 2 is taken away and in its place blackbody 3 is similarly introduced, and signal U_{13} is measured. Immediately after removing blackbody 3, signal U_{11} is again measured. An agreement between this and the earlier measured (zero) values proves the absence of sample heating due to the irradiation by the blackbodies. Next, each blackbody is placed in turn in front of the sample (to the right of the sample in Fig. 2), signals U_{21} and U_{31} are analogously measured, and signal U_{11} is monitored.

Table 1 provides the results of measurements and calculations as applied to a 20-μm-thick film of polyethyleneterephthalate (PETP). In view of the very small scattering of the radiation in the bulk and on the surface of this film, in avoiding its heating the sample is irradiated at less than hemispherical solid angles (for 400-mm blackbody apertures, the blackbodies were placed at distances not less than 200 mm from the sample). We used a larger than necessary number of temperatures for blackbodies 2 and 3 – three values (50, 65, and 80°C) instead of two. This allowed us to conduct measurements and analyze results in three variants (see Table 1).

The cited example shows the fully satisfactory reproducibility of the results of measurements. Thus, the integral coefficients and their deviation limits averaged over all three variants at T_1 = 21°C are $\tau(21) = 0.528 \pm 0.009$, $\rho(21) = 0.132 \pm 0.003$, and $\varepsilon(21) = 0.340 \pm 0.012$; at 80°C are $\tau(80) = 0.484 \pm 0.003$, $\rho(80) = 0.126 \pm 0.001$, and $\varepsilon(80) = 0.390 \pm 0.004$; and the temperature gradients (K^{-1}) are $t = -(7.5 \pm 1.0)\cdot 10^{-4}$, $r = -(0.9 \pm 0.4)\cdot 10^{-4}$, and $e = (8.4 \pm 1.4)\cdot 10^{-4}$. These results confirm the practical feasubility of the proposed measurement method and the validity of the two assumptions used in its development, namely, the first which corresponds to the condition of Eq. (3) and the second to the conditions of Eqs. (16)-(18).

The signals shown in Table 1 were also analyzed using Eqs. (7) and (8), which correspond to the method of [5]. For the conducted measurements, this method is implemented in the specific case of $T_3 = T_1$ (here, $U_3 = U_1$, $U_{32} = U_{12}$, and $U_{23} = U_{21}$). The results of such an analysis and its comparison to measurements are provided in Table 2. These results confirm the conclusions reached during the analysis of Eq. (6) concerning the systematic deviations of the quantities obtained by the methods of [4] and [5] from the measurement results obtained by the present method: in the given case, these deviations are about –10% for

TABLE 2. Comparison of the Results of the Same Series of Measurements Analyzed According to Two Methods

Measured quantities	Method [5]			Present method	
	T_2=50°C	T_2=65°C	T_2=80°C	at 21°C	at 80°C
τ	0,463	0,455	0,448	0,528	0,484
ρ	0,124	0,123	0,122	0,132	0,126
ε	0,413	0,422	0,430	0,340	0,390

τ, about −5% for ρ, and about +15% for ε. This proves that the proposed method significantly corrects the errors in the known methods. It is also proved that the correction is larger for higher absolute values of temperature gradients of the measured quantity.

Summarizing the obtained results, we note that taking into account the temperature dependence of the integral radiation, transmission, and reflection coefficients (which is taken to be linear in the temperature range of the three employed blackbodies) of spectrally selective materials contributes to the lowering of methodical measurement errors that are present in the known methods. With an additional analysis of the measurement results it is possible at the same time to determine the temperature gradients of the sought integral coefficients, which cannot be achieved by other measurement methods.

NOTATION

α, absorption coefficient; ε, emission coefficient; ρ, reflection coefficient; τ, transmission coefficient; σ, Stefan–Boltzmann constant; λ, wavelength; L, irradiance; T, temperature; e, r, t, temperature gradients of the integral emission (e), reflection (r), and transmission (t) coefficients; U, electrical signal at the output of the photodetecting system (PDS). Indices: single indices pertain to blackbodies at the temperatures of o, sample, 1, first (isothermal chamber), 2, second, and 3, third; ij (i, j = 1, 2, 3) refers to the sample under the conditions of a simultaneous irradiation of its visible side by a blackbody at temperature T_i and of its opposite side by a blackbody at temperature T_j.

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THERMAL CONDUCTIVITY OF POLYMER COMPOSITES WITH A DISPERSE FILLER

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UDC 536.2:621.762

A method is proposed for predicting the thermal conductivity of polyolefin composite materials with a disperse filler. The method is based on the combined application of percolation theory and generalized conduction theory. The results of the calculations are in good agreement with experimental data over a wide range of filler concentrations at various temperatures. It is shown that the thermal conductivity of certain composites can be an order of magnitude higher than the thermal conductivity of their matrices.

The thermophysical properties of polymer composites depend on the properties of the basic components: the polymer matrix and the filler. The fillers have superior thermophysical

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Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 62, No. 3, pp. 453-458, March, 1992. Original article submitted July 16, 1991.