# INDIRECT METHODS FOR DETERMINING OPTICAL CHARACTERISTICS OF RADIATION SCATTERING MATERIALS

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An analysis was made of the possibility of using methods for determining the optical characteristics of selectively absorbing and scattering materials based on the measurement of the relative transmission of two samples in single-beam and double-beam spectrophotometers. It is shown that such methods produce large errors and cannot be used in engineering practice. It is recommended that the optical characteristics of such materials be determined by other special methods.

The basis of indirect methods [7, 17, 18] is the determination of the extinction coefficient by measurement of the transmission of two samples of different thickness and the use of the Burger–Lambert law

$$k_\lambda = \frac{1}{l_2 - l_1} \ln \left( \frac{T'_{\lambda,1}}{T'_{\lambda,2}} \right).$$

In Eq. (1), $T'_\lambda$ is understood to be the transmission of a sample measured by the usual technique neglecting scattering. In addition, Equation (1) was obtained under the assumption that reflectivity was independent of layer thickness.

The extinction coefficient obtained from Eq. (1) is subsequently used for calculations of the reflectivity $R_\lambda$ and transmission $T_\lambda$ and also of the degree of darkening $\varepsilon_T$ for a layer of arbitrary thickness from expressions also obtained using the Burger–Lambert law [17, 18].

At the same time, it is well known [3, 6, 8, 12-14, 21] that the Burger–Lambert law is only valid for weakly scattering media with $\beta_\lambda/k_\lambda \ll 0.1$, and can be used in cases of small optical thickness $k_\lambda l < 0.5$ ($T_\lambda > 0.7$) when single scattering is predominant in the layer. In the latter case, for strong scattering, $\beta_\lambda/k_\lambda > 0.9$, the error in the determination of $T_\lambda$ by the Burger–Lambert law without consideration of multiple scattering approaches 10%, but for $k_\lambda l > 1.0$ (i.e., $T_\lambda < 0.5$), the error is even more than 40%, which leads to large error in the calculation of $k_\lambda$ from Eq. (1).

From a solution of the problem of radiation propagation in a plane layer of a scattering and absorbing medium exposed to directed radiation, complicated expressions are obtained for $T_\lambda$ [6, 8, 20, 21, 23] from which it is impossible to determine the value of $k_\lambda$. A simpler approximate expression for the determination of $T_\lambda$ [1, 5, 8, 12, 14] applicable to cases of actual strongly scattering materials and media with characteristic scattering curves strongly peaked forward, which was confirmed experimentally [5, 8, 12, 13], has the form

$$T_\lambda = \frac{\left(1 - R^2_{\lambda\infty}\right) \exp\left(-\sigma_\lambda l\right)}{1 - R^2_{\lambda\infty} \exp\left(-2\sigma_\lambda l\right)}. \tag{2}$$

In this expression $\sigma_\lambda$ and $R_{\lambda\infty}$ depend only on the absorbing and scattering properties of the medium and are determined experimentally [13]. The connection between the parameters $\sigma_\lambda$ and $R_{\lambda\infty}$ and the extinction coefficient $k_\lambda$ and probability of quantum survival $\Lambda = \beta_\lambda/k_\lambda$ is determined from the following equations [1, 8, 14, 21]

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$$\sigma_\lambda = 2k_\lambda \sqrt{(1-\Lambda)(1+\Lambda-2F_\lambda\Lambda)}, \tag{3}$$

$$R_{\lambda\infty} = \frac{1 - F_\lambda\Lambda - \sqrt{(1-\Lambda)(1+\Lambda-2F_\lambda\Lambda)}}{(1-F_\lambda)\Lambda}, \tag{4}$$

where $F_\lambda$ is a parameter which characterizes the degree of peaking in the characteristic scattering curve and is equal to the ratio between the radiation flux scattered into the half-space in the direction of the incident flux and the total flux scattered per unit layer.

The quantity $\sigma_\lambda$, the so-called effective attenuation coefficient [13] or the depth damping factor [8, 20], in contrast to the extinction coefficient $k_\lambda$, characterizes the attenuation of the scattered radiation flux and includes multiple scattering which gives rise to an increase in the intensity of the flux of forward-scattered radiation.

It follows from Eq. (2) that one can determine the effective attenuation coefficient $\sigma_\lambda$, which includes multiple scattering, directly from measurement of the transmission of a material but not $k_\lambda$. This is because the Burger–Lambert law is only a special case of Eq. (2) for $\beta_\lambda/k_\lambda \ll 0.1$ and describes single scattering.

In addition, experimental studies [4, 10] showed that the quantity $T_\lambda$ measured by the usual technique without consideration of scattering is ten times smaller than the true value.

Measurement errors on single-beam and double-beam spectrophotometers depend both on the value of the scattering coefficient of the material and on sample thickness. Samples of different thicknesses are irradiated not by a parallel beam but by a diverging beam of radiation; therefore a decrease in intensity of the direct radiation flux because of beam divergence will be greater for a thicker sample. Multiple scattering of radiation within a sample leads to a condition where additional "direct" radiation scattered by the sample, the intensity of which also depends on sample thickness and the characteristic scattering curve, passes through the spectrometer slit. It is known [6, 8] that radiation scattering in a medium is characterized by broadening of the effective cross section of a narrow beam of radiation; the cross section increases as the optical thickness increases. The effect of an initial angular divergence in the incident beam on the effective cross section of the emerging beam is aggravated by the broadening. Therefore, radiation beams emerging from samples of different thicknesses have not only different spatial distributions but also different effective cross sections.

Only a small portion of the entire radiation flux passing through a sample passes through the slit into the spectrophotometer, and the fractional radiation loss $\Delta T_\lambda$ produced by scattering and broadening of the beam increases disproportionately with increase in sample thickness.

Consequently, it is not possible to eliminate such an error by a choice of sample thicknesses [18]. The relative transmission of two samples is determined with a large, uncontrolled error which gives rise to a considerably larger error in $k_\lambda$ calculated from Eq. (1).

Neglect of reflection losses also has its effect on the resultant value of $k_\lambda$ since in actuality the reflectivity of scattering materials depends on layer thickness and radiation is reflected from all layers, not merely the surface layer [2, 4, 5, 8, 12, 20-23].

Reflection losses are taken into account in the method of [15]; however, the latter was developed only for purely absorbing materials, not scattering materials.

Applying this method [15] to light-scattering materials, one can obtain from Eq. (2) the following formula for determining the effective attenuation coefficient (but not $k_\lambda$) from the relative transmission

$$\sigma_\lambda = \frac{1}{l_2 - l_1}\left[\ln\left(\frac{T_{\lambda,1}}{T_{\lambda,2}}\right) - \Delta D_\lambda\right], \tag{5}$$

where $\Delta D_\lambda$ is a correction which takes into account reflection loss and depends on sample thickness:

$$\Delta D_\lambda = \ln\left[\frac{1 - R_{\lambda\infty}^2 \exp(-2\sigma_\lambda l_1)}{1 - R_{\lambda\infty}^2 \exp(-2\sigma_\lambda l_2)}\right]. \tag{6}$$

Given the ratio of sample thicknesses and the value of $R_{\lambda\infty}$, one can determine $\Delta D_\lambda$ as a function of the observed value of $\ln(T_{\lambda,1}/T_{\lambda,2})$ with scattering included. Consequently, for an analytic determination

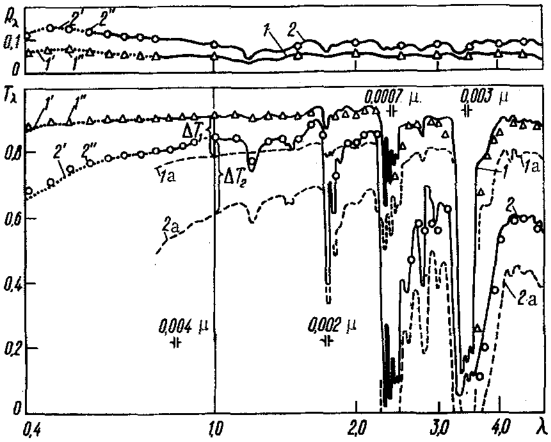


Fig. 1

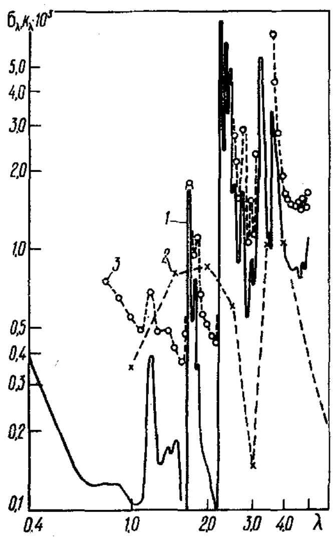


Fig. 2

Fig. 1. Reflectivity $R_\lambda$ and transmission $T_\lambda$ of polyethylene as a function of wavelength $\lambda(\mu)$ and sample thickness (0.05 mm in curves 1, 1', 1", 1a; 0.52 mm in curves 2, 2', 2", 2a) obtained by various methods: 1, 2) double-beam method using mirror hemisphere [11]; 1', 2') integrating sphere method using SF-10; 1", 2") composite method [10] in the spectral region 0.4-1.4 $\mu$ and single-beam method using mirror hemisphere [2, 16] in spectral region 1.0-5.0 $\mu$; 1a, 2a) ordinary double-beam method using IKS-14 and UR-20.

Fig. 2. Dependence of effective attenuation coefficient $\sigma_\lambda$ (1/m) and extinction coefficient $k_\lambda$ (1/m) of polyethylene on wavelength $\lambda$ ($\mu$): 1) $\sigma_\lambda$ calculated from Eq. (8) including scattering and reflection loss; 2) $k_\lambda$ calculated from Eq. (1) [17, 18]; 3) $k_\lambda$ calculated from Eq. (1).

of the quantity $\sigma_\lambda$ from Eq. (5), it is still necessary to know the value of $R_{\lambda\infty}$, which is also determined with scattering included, in addition to the true values of $T_{\lambda,1}$ and $T_{\lambda,2}$. The problem of determining the quantity $\sigma_\lambda$ from two transmission measurements without measurement of $R_{\lambda\infty}$ is only solved graphically [13]. The quantity $k_\lambda$ can be found from Eq. (3) if the parameters $F_\lambda$ and $\Lambda$ are known.

In order to estimate the effect of the deficiencies of the indirect methods enumerated above on the magnitude of the error permissible in calculations of $k_\lambda$ by Eq. (1), we determine the ratio of the transmission $T'_\lambda$ obtained from single-beam and double-beam spectrophotometers for two samples of different thicknesses $l_2 > l_1$. Assuming the material scatters radiation weakly, $\Lambda \ll 0.1$, but considering that the reflectivity depends on layer thickness, we obtain the following expression by using the Burger–Lambert law:

$$\frac{T'_{\lambda,1}}{T'_{\lambda,2}} = \left(\frac{1-R_{\lambda,1}}{1-R_{\lambda,2}}\right) \exp\left[k_\lambda (l_2 - l_1)\right], \tag{7}$$

where

$$T'_{\lambda,1} = T_{\lambda,1} - \Delta T_{\lambda,1}; \; T'_{\lambda,2} = T_{\lambda,2} - \Delta T_{\lambda,2}.$$

One can then write for the determination of $k_\lambda$ including scattering and reflection loss

$$\sigma_\lambda \simeq k_\lambda = \frac{1}{l_2 - l_1} \ln\left[\left(\frac{1-R_{\lambda,2}}{1-R_{\lambda,1}}\right)\left(\frac{T'_{\lambda,1} + \Delta T_{\lambda,1}}{T'_{\lambda,2} + \Delta T_{\lambda,2}}\right)\right]. \tag{8}$$

A comparison of Eqs. (1) and (8) indicates that the values of $k_\lambda$ for various materials obtained in [7, 17, 18] were overestimated even in the cases where the Burger–Lambert law was applicable for the following two reasons: 1) the quantity $(1-R_{\lambda,2})/(1-R_{\lambda,1})$ is always less than one since $R_{\lambda,2} > R_{\lambda,1}$; 2) the quantity $\Delta T_{\lambda,2}$ is always greater than $\Delta T_{\lambda,1}$ because the broadening of the cross section of the radiation beam in the thicker sample $l_2$ is considerably more than in the thinner sample $l_1$. It then follows that $(T'_{\lambda,1}/T'_{\lambda,2}) > (T_{\lambda,1}/T_{\lambda,2})$.

Because $R_\lambda$ and $T_\lambda$ appear in the logarithmic term in Eq. (8), an error of 5-10% in the determination of these quantities leads to an error of 200-600% in the calculation if $k_\lambda$. The error in measurement of relative transmission associated with scattering and broadening of the radiation beam in the samples can be reduced by a reduction in the difference in sample thicknesses. In this case, however, the value of the ratio between $T_{\lambda,1}$ and $T_{\lambda,2}$ will be close to one which gives rise to a sharp increase in the error of calculations for arbitrary $T_{\lambda,2}$.

To arrive at a numerical estimate of the error introduced into the value of $k_\lambda$ determined by Eq. (1) for selectively absorbing and scattering materials, the quantities $R_\lambda$ and $T_\lambda$ were measured (Fig. 1) for two polyethylene samples, including scattering, on the basis of which values of $\sigma_\lambda$ and $k_\lambda$ were calculated from Eqs. (1) and (8) (Fig. 2). Sample thicknesses were chosen so that the transmission of the thicker sample $T_{\lambda,2}$ was greater than 0.7 for $l_1/l_2 < 0.1$, which made it possible to use the Burger–Lambert law with small error because single scattering predominates in such a layer thickness.

Measurements of $R_\lambda$ and $T_\lambda$ for polyethylene samples 0.05 and 0.52 mm thick were carried out in the spectral region 0.4-1.4 $\mu$ by a composite method (points 1", 2"), and in the spectral region 1.0-5.0 $\mu$ by a double-beam method (curves 1, 2) with the help of special attachments to the spectrophotometers that made it possible to include radiation scattering by the samples [10, 11]. The results were checked in the spectral region 0.40-0.75 $\mu$ by the integrating sphere method using an SF-10 (points 1', 2') and in the spectral region 1.0-5.0 $\mu$ using an IKS-12 with a hemispherical attachment [2, 11, 16] (points 1", 2"). As is clear from Fig. 1, there is good convergence of the results for $R_\lambda$ and $T_\lambda$ measured by the methods mentioned.

Curves 1a and 2a (Fig. 1) are experimental confirmation that polyethylene scatters radiation strongly and that the value of $\Delta T_{\lambda,2}$ is indeed greater than that for $\Delta T_{\lambda,1}$. Thus $\Delta T_{\lambda,2}$ is twice as great as $\Delta T_{\lambda,1}$ at a wavelength of 1.0$\mu$. Therefore the observed value, without including scattering, of the quantity $\ln(T'_{\lambda,1}/T'_{\lambda,2}) = 0.2546$ is 5.02 times greater than the true value, which is 0.0488.

The reflectivity of polyethylene depends on layer thickness. For a sample thickness of 0.52 mm, the value of $R_{\lambda,2}$ varies from 0.047 to 0.135 and for a thickness of 0.05 mm, $R_{\lambda,1} = 0.034$-0.065 in the spectral region 0.4-5.0 $\mu$, i.e., $R_{\lambda,2} > R_{\lambda,1}$. For a wavelength of 1.0 $\mu$, therefore, the value of the ratio $(1-R_{\lambda,2})/(1-R_{\lambda,1})$ is 0.965 and the value of $k_\lambda$ calculated without including reflection but including scattering is an overestimate by a factor of 1.57 in comparison with the true value.

From a comparison of the results presented in Fig. 2 for calculations of $\sigma_\lambda$ from Eq. (8) and $k_\lambda$ from Eq. (1), it follows that losses in reflection and scattering do indeed lead to an overestimate of the values of $k_\lambda$ (curve 3).

Values of $k_\lambda$ (curve 2) given in [17, 18] were overestimated by a factor of 3.35 at $\lambda = 1.0\ \mu$, by a factor of 4.37 at $\lambda = 1.5\ \mu$, and by a factor of 5.90 at $\lambda = 2.0\ \mu$. In the spectral region 2.2-5.0 $\mu$, values of $k_\lambda$ were underestimated by factors of 2.7-5.6, which is explained by the large error in measurement of the quantity $T'_{\lambda,1}/T'_{\lambda,2}$ since for the polyethylene samples 0.820 and 1.630 mm thick used for the study [17] in this spectral region, the transmission $T_{\lambda,2}$, measured by the usual methods, was less than 20% and fell to 0% over broad wavelength ranges of 2.27-2.78 $\mu$ and 3.12-4.50 $\mu$. Consequently, values of the quantities $R_\lambda$ and $\varepsilon_\lambda$ for polyethylene given in [17, 18] were also determined with a large error.

For paint and varnish materials and coatings, the error in the determinations of $k_\lambda$, $R_\lambda$, and $\varepsilon_\lambda$ given in [17, 18] is greater for greater optical density of the material studied and when radiation scattering is stronger. Studies performed [2] showed that paint and varnish materials and coatings scatter radiation strongly.

Thus indirect methods for determining the optical characteristics of radiation scattering materials based on measurements of relative transmission by the usual techniques using single-beam and double-beam spectrophotometers yield large errors (200-600%) and cannot be used for practical purposes.

It is impossible to determine the extinction coefficient from measurements of the transmission of two samples of scattering materials of different thickness.

Data on $k_\lambda$, $R_\lambda$, and $\varepsilon_\lambda$ for paint and varnish materials and coatings, for polyethylene Teflon, and for porous-capillary colloidal materials, obtained by indirect methods [7, 17, 18] were determined with large errors and cannot be used in engineering calculations.

The optical and thermoradiative characteristics of materials that absorb and scatter radiation should be determined by special methods [2, 8-14, 16, 22].

## NOTATION

$\alpha_\lambda$, $\beta_\lambda$, $\sigma_\lambda$, $k_\lambda$ are the spectral coefficients of absorption, dissipation, effective attenuation, and extinction, $m^{-1}$;

$R_\lambda$, $T_\lambda$, $A_\lambda$ are the reflective, transmitting, and absorbing powers of a flat layer of thickness $l$;

$R_{\lambda\infty}$ reflectivity of optically infinitely thick layer.

## LITERATURE CITED


1. V. A. Ambartsumyan, Scientific Works [in Russian], Vol. 1, Izd. AN ArmSSR (1960).
2. G. D. Afanas'eva, L. M. Vinogradova, S. G. Il'yasov, M. B. Fridzon, and B. F. Tyurin, Lakokrasochnye Materialy i ikh Primenenie, No. 4, 38 (1969).
3. S. I. Vavilov, Collected Works [in Russian], Vol. 1 (1954).
4. A. S. Ginzburg, V. V. Krasnikov, and N. G. Selyukov, Inzh.-Fiz. Zh., 8, No. 6, 742 (1965).
5. M. M. Gurevich, Introduction to Photometry [in Russian], Energiya, Leningrad (1968).
6. L. S. Dolin, Izv. VUZ. Radiofiz., 9, No. 1, 61 (1966).
7. V. P. Dushchenko, A. F. Bulyandra, and I. M. Kucheruk, Izv. VUZ. Pishchevaya Tekhnologiya, No. 4, 152 (1967).
8. A. P. Ivanov, Optics of Scattering Media [in Russian], Nauka i Tekhnika, Minsk (1969).
9. G. K. Il'ich, Zh. Prikl. Spektroskopii, 4, No. 5, 459 (1966).
10. S. G. Il'yasov, V. V. Krasnikov, M. B. Fridzon, and V. I. Shlyakhov, Meteorologiya i Gridrologiya, No. 10, 96 (1968).
11. S. G. Il'yasov, Abstracts of Papers at All-Union Young Scientists Conference on Heat and Mass Exchange [in Russian], Minsk (1969), p. 177; Inzh.-Fiz. Zh., 18, No. 4 (1970).
12. S. G. Il'yasov and V. V. Krasnikov, Inzh.-Fiz. Zh., 15, No. 2 (1968).
13. S. G. Il'yasov and V. V. Krasnikov, Inzh. Fiz. Zh., 17, No. 2 (1969).
14. S. G. Il'yasov, in: Proceedings of the Moscow Technical Institute of the Food Industry: Processes and Equipment in Food Production [in Russian], Moscow (1969), p. 49.
15. L. D. Kislovskii and R. I. Kurtsinovskaya, Zh. Prikl. Spektroskopii, 8, No. 3, 489 (1968).
16. M. A. Kropotkin and B. P. Kozyrev, Opt. Spektrosk., 17, No. 2, 259 (1964).
17. G. D. Rabinovich, Inzh.-Fiz. Zh., 5, No. 9, 16 (1962).
18. G. D. Rabinovich and L. S. Slobodkin, Thermoradiative and Convective Drying of Paint and Varnish Coatings [in Russian], Nauka i Tekhnika, Minsk (1966).
19. G. D. Rabinovich and L. S. Slobodkin, in: Problems of Thermoradiative Technology and Drying of Paint and Varnish Coatings [in Russian], Minsk (1969).
20. G. V. Rozenberg, Dokl. Akad. Nauk SSSR, 145, No. 4, 775 (1962).
21. A. M. Samson, Dokl. Akad. Nauk, BSSR, 3, No. 5, 197 (1959).
22. B. I. Stepanov, Yu. I. Chekalinskaya, and O. P. Girin, Trudy Instituta Fiziki i Matematiki, Akad. Nauk Belorussian SSR, 1, 152 (1956).
23. S. Chandresekar, Transport of Radiant Energy [Russian translation], IL (1953).