Solving the inverse problems on equivalent classes

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We present an approach to solving inverse problems based on classification of isotropic ensembles of non-spherical particles. The hypothesis that equivalent ensembles have close extinction coefficients is formulated and checked. The results are illustrated by numerical calculations of extinction coefficients for randomly oriented ellipsoidal particles, hexagonal and circular cylinders. The absorption coefficients for the pigment mixture of *Spirulina platensis* within the photosynthetic band (λ = 410 to 700 nm) are estimated. The degree of destruction of chloroplasts under mechanical effect is calculated.

The variety of shapes of non-spherical particles, even without the account of their internal structure makes it impossible to solve the inverse problems on determining the microstructure (size and shape distributions) of the ensembles of particles.

In this paper we consider the approach, which is based on the classification of ensembles of non-spherical convex particles upon microstructure parameters. The isotropic ensembles of particles are related to the same class if they have three equal moments of the distribution – the ensemble average surface $<S>$, volume $<V>$, and the squared volume $<V^2>$, respectively, the second, third, and the sixth moments of the distribution. The working hypothesis is considered and examined, that the isotropic ensembles of particles belonging to the same class of equivalence have close values of the extinction, scattering, and absorption coefficients, the relative error being used as a criterion.

The basis for the statement of the working hypothesis is the optical equivalence, proved in Refs. 1 and 2 (in the Rayleigh–Gans–Debye and anomalous diffraction approximation[3]), of the polydisperse ensembles of randomly oriented ellipsoids, spheroids, and spherical particles, which have three aforementioned moments of the distribution. An ensemble of spherical particles with power size distribution[2] is selected as a representative of the class of equivalence the optical characteristics of which are the estimates of those for each representative of this class. Thus, solution of the inverse problem can be reduced to the solution on the class of equivalence and determination of the parameters of the power-law size distribution in the range where the hypothesis has been obtained.

Correct application of this approach is possible if the following necessary conditions have been fulfilled: a) single scattering, b) the absence of physical fields orienting the particles, or their resultant is equal to zero, c) the shape of particles is assumed to be convex, d) the ensemble of particles is isotropic and includes the randomly oriented particles and their mirror reflections relative to the plane of scattering.

Variations of the extinction coefficients are estimated in the ranges of the classes of equivalence containing ellipsoid particles, finite round and hexagonal cylinders. The refractive indices of the mixture of pigments in the visible wavelength range are estimated using the experimental data on the optical spectra of absorption of the aquatic plant *Spirulina platensis*.

1. Optical equivalence of isotropic ensembles of ellipsoid particles

As was shown[1,2] the extinction, scattering, and absorption coefficients C of randomly oriented ellipsoidal particles in the Rayleigh–Gans–Debye and anomalous diffraction approximation are equal to:

1) the corresponding coefficients of three different (due to permutation of a, b, and c) polydisperse ensembles of randomly oriented spheroidal particles

$$< C(a,b,c) >= \frac{2a^2b^2}{\pi}\int\limits_a^b d\hat{a} < C(\hat{a},\hat{a},c) > \frac{\hat{a}^{-3}}{\sqrt{(b^2-\hat{a}^2)(\hat{a}^2-a^2)}}, \quad (1)$$

where a, b, and c are the half-axes of an ellipsoidal particle;

2) the light scattering coefficients of spherical particles with the weighting function invariant with respect to permutations of the a, b and c:

$$\rho_{eq}(r) = \Theta(c-r)\Theta(r-a)\frac{2a^2b^2c^2}{\pi r^5}\times \int\limits_a^{\min(r,b)} d\hat{a}\frac{\hat{a}}{\sqrt{-(b^2-\hat{a}^2)(a^2-\hat{a}^2)(c^2-\hat{a}^2)(r^2-\hat{a}^2)}}, \quad (2)$$

here $\Theta(x)$ is the Heaviside function.

After the corresponding substitution $x = \hat{a}^2$ the integral obtained is reduced to the full elliptical integral of the 1st kind in the Legendre form.[2]

The proved optical equivalence has a wider range of applicability than the ranges of correct application of the Rayleigh–Gans–Debye ($|m_r - 1| \ll 1$,

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$2kr|m_r - 1| \ll 1$) and anomalous diffraction ($|m_r - 1| \ll 1$, $kr \gg 1$) approximations; here m_r is the relative refractive index of the particulate mater, r is the characteristic size of particles, $k = 2\pi/\lambda$ is the wave number, and λ is the wavelength of the incident radiation. The dependence of the maximum relative error of calculated extinction coefficients in pairs among four equivalent polydisperse ensembles of particles and spherical particles with the power-law size distribution (see Section 2) on m_r and kc ($a : b : c = 1 : 2 : 3$) shown in Fig. 1 is an evidence of this fact. These ensembles of particles have three equal moments of distribution $<S>$, $<V>$, $<V^2>$. The kernel of the integral operator in Eqs. (1) and (2) was calculated using the exact methods: the method of T-matrices and Mie theory.

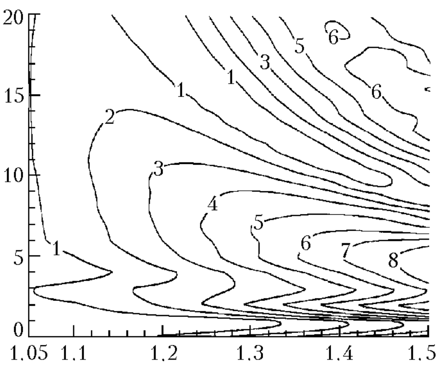

Fig. 1. Maximum relative error (%) of the extinction coefficients in pairs among equivalent ensembles of randomly oriented ellipsoidal particles (1), (2) and the equivalent polydisperse ensemble of spherical particles with the power-law distribution as a function of the maximum diffraction parameter kc at different relative refractive indices of the particulate matter.

The logic continuation of the optical equivalence is the classification of isotropic ensembles of non-spherical particles, which is considered in the next section.

2. Classification of isotropic ensembles of homogeneous non-spherical particles

The equality of three microstructure parameters is the equivalence condition and it divides all isotropic ensembles into the classes of equivalence. Let us call isotropic ensembles of particles belonging to the same class equivalent. Let us use a polydisperse ensemble of spherical particles with the power-law size distribution and the distribution density function of the following form as a representative characterizing the class:

$$f(r) = \begin{cases} c r^{-5}, & r_{\min} \le r \le r_{\max} \\ 0, & r \notin [r_{\min}, r_{\max}] \end{cases}.$$

The parameters of the distribution c, $r_{\min}$, $r_{\max}$ can be determined in the explicit form from the system of equations:

$$\begin{cases} c\pi \int_{r_{\min}}^{r_{\max}} r^{-3} \mathrm{d}r = < S >, \\ c\dfrac{4\pi}{3} \int_{r_{\min}}^{r_{\max}} r^{-2} \mathrm{d}r = < V >, \\ c\dfrac{16\pi^2}{9} \int_{r_{\min}}^{r_{\max}} r \mathrm{d}r = < V^2 > . \end{cases}$$

The comparison of the extinction coefficients of polydisperse ensembles of randomly oriented finite round cylinders and spherical particles with the power-law size distribution are shown in Fig. 2 as functions of the relative refractive index of the particulate matter.

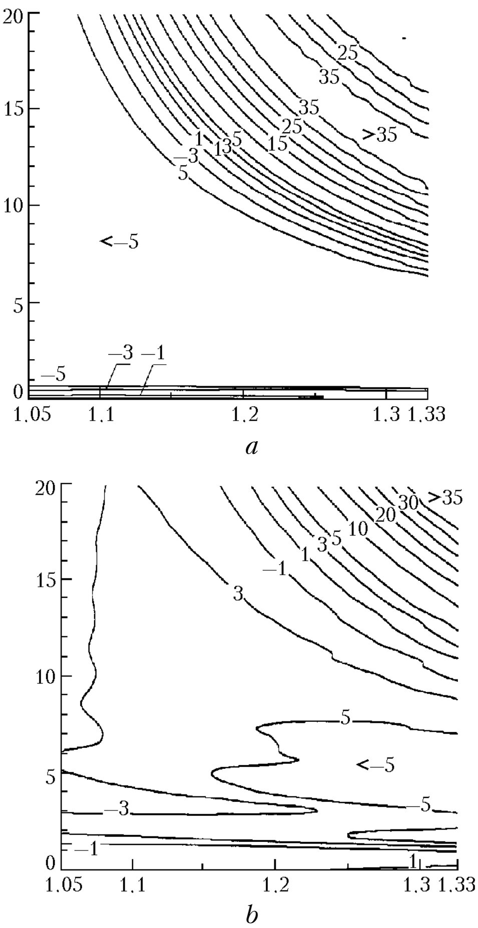

Fig. 2. Relative error Δ (%) in calculating the extinction coefficients of the ensemble of randomly oriented oblate ($\varepsilon = 2$, *a*) and elongated ($\varepsilon = 0.5$, *b*) round cylinders by means of the equivalent ensemble of spherical particles as a function of the relative refractive index m_r and the maximum diffraction parameter $\rho_{\max}$; ε is the ratio of the base diameter to the length of the cylinder.

The calculations were performed using the exact methods. The relative error was calculated by the formula

$$\Delta = \frac{C_{\text{ext}}^{\text{a}} - C_{\text{ext}}^{\text{e}}}{C_{\text{ext}}^{\text{e}}} 100\%,$$

where the superscript "a" corresponds to calculations of the extinction coefficient using the equivalent ensemble of spherical particles, and "e" corresponds to the exact calculations using the method of T-matrices.

The hypothesis is also confirmed for the hexagonal cylinders with the ratio of the length L to the diameter d of 6:1 and the relative refractive index of $1.311 + 3.11 \cdot 10^{-9} i$. The relative error in calculating the efficiency factors of extinction of the randomly oriented hexagonal cylinders by means of the FDTD method (Finite Difference Time Domain Method)[4] and the equivalent ensemble of spherical particles with the power-law size distribution does not exceed 10% in the range $6 \le kL \le 20$ (Fig. 3).

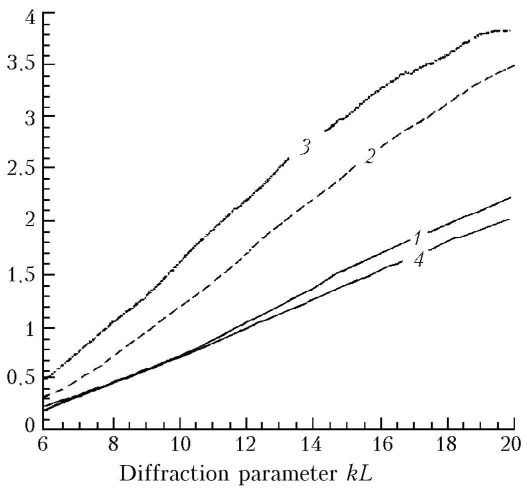

Fig. 3. The extinction efficiency factor of randomly oriented ice hexagonal cylinders ($L/d = 6$) calculated using the FDTD method (*1*), equivolume (*2*) and equisurface (*3*) spherical particles[4] and by means of the equivalent ensemble of spherical particles (*4*).

The absorption coefficient is the most conservative characteristics among the light scattering coefficients, and the simplifications are possible for the optically "soft" particles – the classes of equivalence are determined by the equality of the second and third moments of the distribution. In this case the representative of the class of equivalence is a single particle of a cylindrical shape, whose axis of symmetry is oriented along the direction of radiation incidence. The volume and the area of projection of this particle coincide with the analogous parameters of the suspension of particles under study, and the absorption coefficient is calculated using the anomalous diffraction approximation.

3. Experimental analysis of the optical absorption spectra of aqua plants (on the example of *Spirulina platensis*)

The estimate of the absorption coefficient of isotropic biological suspension consisting of randomly oriented cells of the volume V and the geometric cross section S has, in the single scattering approximation, the following form[5]:

$$C_{abs}(\lambda) = [1 - \exp(-\alpha(\lambda)\nu)]S, \tag{3}$$

where $\nu = \langle V \rangle / \langle S \rangle$, $\langle V \rangle$, and $\langle S \rangle$ are, respectively, the volume and the geometric cross section averaged over the suspension of cells; $\alpha = 4\pi\chi/\lambda$ is the absorption index, λ is the wavelength of the incident radiation; χ is the imaginary part of the refractive index of the particulate matter $m_r = n_r + i\chi$.

Using Eq. (3), one can estimate transformation of the absorption spectra at a constant mass of the substance (or V). The formula for the transformation coefficient equal to the ratio of the absorption coefficients of the suspension of cells and the solution of the cell substance has the form[5,6]:

$$\eta = \left(1 - \exp(-\alpha\nu)\right)/\alpha\nu \tag{4}$$

and characterizes the specific absorption coefficient as a function of the suspension disperse composition $\alpha\eta = C_{abs}/V$.

If the microstructure of the suspension of cells has been known and the absorption spectra of the suspension of cells and the solution have been measured, one can, using Eq. (4), estimate the spectrum of the absorption indices of the substance in the wavelength range considered. However, there are some difficulties in obtaining solutions of the pigments, because no universal solvent exists for the entire set of pigments, which dissolves some of them and does not destroy the others.

According to Eq. (3), the ratio of the absorption coefficients of the suspensions different in only the geometric cross section S, has the form

$$k(\lambda) = \left[1 - \exp\left(-\alpha(\lambda)\nu_1\right)\right]\nu_2 / \left[1 - \exp\left(-\alpha(\lambda)\nu_2\right)\right]\nu_1. \tag{5}$$

In order to estimate the absorption indices $\alpha(\lambda)$ of the mixture of pigments of the aqua plant *Spirulina platensis* using Eq. (5), the suspension of cells underwent the mechanical influence by means of the ultrasound disintegrator at a frequency of 1MHz that led to the change of S, but the volume V remained the same. The data on the absorption spectra of two aforementioned samples were obtained by means of the spectrophotometer SF-14 and are shown in Fig. 4. In measuring, the condition of single scattering necessary for the correct application of Eq. (3), was fulfilled due to a decrease of the number density of cells.

The absorption indices $\alpha(\lambda)$ at the known (measured) ν_i, $i = 1, 2$, and measured $k(\lambda)$ are the solution of the non-linear equation (5), which is solved by the method of simple iterations. Analysis of the optical absorption spectra has shown that the pigments which have the absorption maximum in the range $410 \le \lambda \le 450$ nm are less subject to the aforementioned mechanical influence. All subsequent calculations were performed assuming that the pigments at $\lambda = 415$ nm do not lose the property of absorbing light after disintegration of the cells.

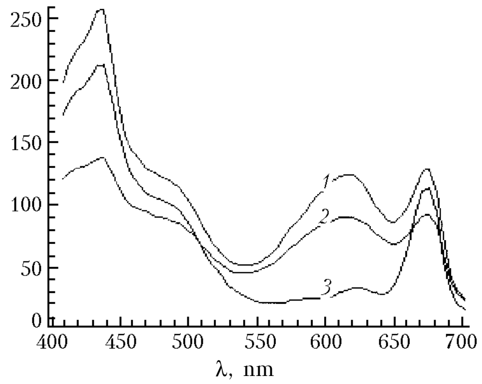

Fig. 4. Optical absorption spectra (in conditional units of the optical density) of native (living) cells of blue-green aqua plant *Spirulina platensis* (*2*), the cells after ultrasound influence (*3*), and the spectrum of the pigment mixture solution reconstructed by means of calculations (*1*).

Microscopic measurements of the microstructure parameters of a sample of native (living) cells of the aqua plant corresponded to $\nu = 5.73$ μm, and that of the sample that has undergone the ultrasonic treatment corresponded to $\nu = 1.61$ μm. The estimates of the microstructure parameters are point-wise being obtained based on the data sample with a cylindrical particle used as the cell model.

The method of determining the absorption index spectra $\alpha(\lambda)$ assumes a number of stages:

a) determination of $\alpha(415)$ from Eq. (5) using known data of microscopic analysis ν_i, $i = 1, 2$, and the known value $k(415)$;

b) determination of $\alpha(\lambda)$ from the measured ratio (see Fig. 4)

$$\beta(\lambda)=\left[1-\exp\left(-\alpha(\lambda)\nu\right)\right]/\left[1-\exp\left(-\alpha(415)\nu\right)\right] \quad (6)$$

for the native cells of the aqua plant ($\nu = 5.73$ μm).

The estimates of $\alpha(\lambda)$ and $\chi(\lambda)$ are presented in the Table.

The absorption spectrum of the mixture of pigments was reconstructed by means of calculations based on the known $\alpha(\lambda)$, the results are shown in Fig. 4.

The known $\alpha(\lambda)$ allows one to determine the microstructure parameter ν of the biological suspension of the cells from the measurement data with the subsequent reconstruction by Eq. (4) of the absorption index spectra of the suspension of pigments. It is known[5,6] that the absorption coefficient of the solution of pigments ($\nu \ll 1$) is directly proportional to the volume (biomass) $C_{\text{abs}}(\lambda) = \alpha(\lambda)V$. The use of this fact, as well as a comparison of the reconstructed absorption spectra of solutions allow one to estimate the relative value of biomass of the aqua plants, and its absolute value, if the biomass of one of the samples has been known.

The absorption coefficient of the suspension of biological cells is a monotonically decreasing function of ν. Disintegration of the cells is accompanied by the decrease of ν. However, in the range $510 < \lambda < 660$ nm the situation is the opposite to the expected one, that is related to the destruction of special structure units, chloroplasts, where the pigments interacting between each other and with other substances of the cell are located, and, hence, the decrease of the absorption ability is observed. Let us indirectly estimate the

Absorption indices of the mixture of pigments of the aqua plant *Spirulina platensis*

λ, nm	$\alpha(\lambda)$, μm^{-1}	$\chi(\lambda)\cdot 10^3$	γ, %
410	0.197	6.42	0.3
415	0.212	7.01	0.0
420	0.222	7.43	0.2
425	0.230	7.76	0.3
430	0.241	8.24	0.8
435	0.255	8.82	1.2
440	0.257	8.99	1.4
445	0.216	7.64	0.9
450	0.180	6.43	1.0
455	0.155	5.59	1.2
460	0.143	5.24	4.4
465	0.135	4.98	7.2
470	0.130	4.88	7.9
475	0.124	4.69	7.8
480	0.121	4.60	8.0
485	0.119	4.58	8.5
490	0.115	4.48	8.5
495	0.111	4.38	10.6
500	0.104	4.14	12.0
505	0.094	3.77	14.2
510	0.083	3.35	17.1
515	0.074	3.04	22.2
520	0.066	2.74	24.0
525	0.059	2.48	27.4
530	0.054	2.28	35.5
535	0.051	2.19	40.2
540	0.050	2.15	46.7
545	0.051	2.23	52.0
550	0.053	2.30	58.9
555	0.055	2.44	62.7
560	0.059	2.64	66.9
565	0.063	2.85	69.0
570	0.072	3.27	71.3
575	0.079	3.64	72.1
580	0.086	3.95	73.6
585	0.092	4.29	74.3
590	0.101	4.72	76.5
595	0.108	5.09	78.0
600	0.111	5.31	78.7
605	0.117	5.62	78.0
610	0.121	5.85	76.2
615	0.122	5.99	74.9
620	0.122	6.04	73.3
625	0.117	5.81	71.9
630	0.111	5.57	71.4
635	0.101	5.08	70.4
640	0.092	4.69	69.9
645	0.086	4.40	66.5
650	0.084	4.35	57.4
655	0.089	4.63	44.5
660	0.099	5.19	28.5
665	0.113	5.98	12.0
670	0.124	6.63	3.4
675	0.127	6.84	3.8
680	0.109	5.92	9.1
685	0.073	4.00	22.9
690	0.044	2.40	25.1
695	0.031	1.69	32.5
700	0.024	1.33	34.1

degree of destruction of the pigments γ (%) by estimating the decrease in the absorption

$$\gamma = 100\left[\alpha(\lambda) - \alpha^{D}(\lambda)\right] / \alpha(\lambda),$$

where $\alpha^{D}(\lambda)$ is the absorption index calculated on the basis of the absorption spectrum of the suspension of cells after the ultrasonic treatment. The decrease of absorption is interpreted as the decrease of the concentration of pigments. The data on the degree of destruction of the pigments are shown in the Table.

Let us note that the estimates of the absorption indices are point-wise and depend on: 1) the choice of the formula for calculation of the absorption coefficients, 2) the estimates of the microstructure parameters based on the sample, and 3) the error in measuring the optical density. These facts introduce an error into determining the absorption indices, their effect on the final result is not considered here and requires special study.

The obtained values α(λ) of *Spirulina platensis* agree with analogous data for the blue-green aqua plant *Chroococcus sp.*[7]

Having known the absorption indices of the pigments in the visible wavelength range, one can estimate the concentration (biomass) of the monoculture of aqua plants, as well as investigate the change of the pigment composition of aqua plants depending on the external conditions (light, nutrition) and physiological state of the cells.

Conclusion

One can consider the approach stated in the paper as the first approximation to solving the inverse problems. Given *a priori* data on the particle shape and the refractive index the solution procedure includes the following stages.

1. Determination of the range of variations of the parameters (size, shape, and refractive index of particles) where the hypothesis is accepted. The criterion is the relative error in calculating the optical characteristics using the polydisperse ensemble of equivalent spherical particles.

2. Solution of the inverse problem in the range of acceptance of the hypothesis is reduced to determining the parameters of the equivalent ensemble with the power distribution c, $r_{\min}$, $r_{\max}$.

3. If $r_{\min}$, $r_{\max}$ have been known, the following relative values are determined: $<V>/<S>$, $<V^2>/<S>$, characterizing the distribution of the relative volume of particles.

Let us note that the present approach is the most effective in investigating the suspensions of biological particles.

Acknowledgments

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