Remote laser diagnostics of polymodal aerosol particles

V.A. Arkhipov, S.S. Bondarchuk, E.A. Kozlov, and V.F. Trofimov

Scientific Research Institute of Applied Mathematics and Mechanics at the Tomsk State University

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Two methods for laser diagnostics of size spectra and concentrations of polymodal aerosol particles are proposed. These methods are modifications of the well-known small-angle scattering diagram method and the Shifrin's spectral transparency method. The corresponding inverse problems of aerosol optics are formulated and solved by the direct search technique. The proposed methods are verified by solving model problems.

The size and concentration of particles in supersonic two-phase plasma flows of combustion products are among the most important parameters determining power and ecological characteristics of various devices (MHD generators, jet propulsions, gas-dynamic test units, etc.). Application of mechanic samplers for determination of particle sizes disturbs the studied flow and seems rather problematic in high-temperature conditions. The existing methods of optical diagnostics of aerosol parameters[1] are usually applicable to unimodal particle size distributions, though the results of some investigations evidence the presence of bimodal and polymodal particle size spectra in the collected samples (in particular, for condensed combustion products of solid rocket propellant[2]). Besides, application of classic methods of optical diagnostics in this case is difficult because of the high-power radiation emitted by plasma and the effects of dynamic lag of particles.

In this paper, we consider two methods for laser diagnostics of polymodal particle size distributions. These methods are modifications of the small-angle scattering diagram method and the spectral transparency method developed by Shifrin.[3] The rights to these methods and the technique for calculation of optical characteristics of polydisperse aerosol systems are protected by the inventor's certificate.[4]

1. Modified small-angle scattering diagram method

Analysis of scattering phase functions for actual polydisperse systems shows that the scattered radiation intensity $I(\Theta)$ peaks within the scattering angle range $\Theta \approx 7{-}10°$ (Ref. 5). The behavior of the scattering phase function in this range (nonmonotonicity, inflection points, large gradients) is largely determined by the polymodal character of the sought particle size distribution functions $f(r)$. The reliable, physically adequate interpolation and extrapolation of $I(\Theta)$ is problematic at a limited number of experimental observations (especially, near $\Theta \approx 0°$, where measurements are technically difficult). The use of regularization algorithms traditionally employed for solving of this class of inverse problems does not provide the acceptable accuracy, and in the case of the polymodal distribution $f(r)$ the solution can hardly be found.[6,7]

To solve this problem, we propose a method and an algorithm based on the direct search method. The solution uniqueness in this case is achieved by using some *a priori* information about the solution, in particular, the number of modes (peaks) of the sought distribution function $f(r)$. In this case $f(r)$ is represented as a series

$$f(r) = \sum_{i=1}^{M} a_i f_i(r), \quad \sum_{i=1}^{M} a_i = 1, \tag{1}$$

where $f_i(r)$ is a unimodal distribution function of some particular form; a_i is the weighting coefficient of the ith function.

At the sufficient number of terms M, the serial representation (1) describes the actual distributions with the preset accuracy. The polydisperse scattering phase function for every particular function $f(r)$ is determined by the equation

$$I(\Theta) = \int_0^{\infty} \pi Q_s(r,\Theta) r^2 f(r)\,\mathrm{d}r, \tag{2}$$

where $Q_s(r,\Theta)$ is the scattering efficiency factor for a particle with radius r.

In the small-angle range, the kernel of the integral equation (2) on the assumption of spherical particles has a simple analytic form[1,3]:

$$Q_s(r,\Theta) = \frac{\rho^2}{4\pi} F(z),$$

$$F(z) = \left[\frac{2J_1(z)}{z}\right]^2, \quad \rho = \frac{2\pi r}{\lambda}, \quad z = \Theta\rho. \tag{3}$$

Here ρ is the diffraction parameter (Mie parameter); λ is the sensing radiation wavelength; $J_1(z)$ is the first-kind first-order Bessel function.

It follows from Eqs. (1)–(3) that:

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$$I(\Theta)=\sum_{i=1}^{M}a_i\int_0^{\infty}\left[\frac{rJ_1(z)}{\Theta}\right]^2 f_i(r)\,\mathrm{d}r=\sum_{i=1}^{M}I_i(\Theta),$$

where $I_i(\Theta)$ is the polydisperse scattering phase function for the ith distribution function $f_i(r)$.

Determination of $f(r)$ from the measured scattering phase function $I(\Theta)$ through solution of direct problems can be reduced to minimization of the functional

$$\wp=\sum_{j=1}^{\Im}\left|I(\Theta_j)-\sum_{i=1}^{M}a_iI_i(\Theta_j)\right|, \qquad (4)$$

where $I(\Theta_j)$ (j = 1, 2, ..., $\Im$) are the measured values of the scattering phase function for discrete values of the scattering angle Θ_j.

The algorithm for problem solving includes several stages. For definiteness, we specify a set of the generalized gamma-distributions[8]:

$$f_i(r) = b_i\, r^{\beta_i} \exp[-(r/r_{0i})^2], \qquad (5)$$

as the system of the functions $f_i(r)$. Here r_0 is the maximum (mode) of the distribution function; b and β are the distribution parameters.

At the preliminary stage, we compile a table of the calculated values of $I_i(\Theta_j)$ for the measurement angles Θ_j and measurement limits of the modal radius r_0 selected based on physical reasons (in the small-angle method, minimal r_0 is the wavelength λ).

At the first approximation stage, varying the weight coefficients $a_i = 0, \frac{1}{M}, \frac{2}{M}, \ldots, \frac{M-1}{M}, 1$ (where M is the degree of "splitting"), we determine their set providing for the minimum of the functional (4). Variants $\{a_i\}$ are generated with allowance for the restriction imposed by the *a priori* preset number of modes of the sought distribution function. For the unimodal distribution this restriction is formulated as a generation $\{a_i\}$ with one maximum, for bimodal distributions – as a generation with two maxima, and so on.

The simplest refinement of the solution at the second and following stages is varying of each coefficient a_i within $1/M$ about the pre-determined value for further minimization of the functional (4). Another possible method to refine $f(r)$ is the use of the information obtained at some stage of varying in order to approximate the experimental values of the scattering phase function by some function and then to solve the inverse problem.

The results of software tests are shown in Figs. 1 and 2. The figures depict the dimensionless distribution functions $\bar{f}(r)=f(r)/f(r_0)$, where r_0 is the modal radius of particles of the initial size. In the both cases, we specified four values of the scattering phase function for the angles 0.5, 2, 5, and 10°. The solution was sought as a sum of a five-term series of unimodal functions of the generalized gamma-distribution having the form (5).

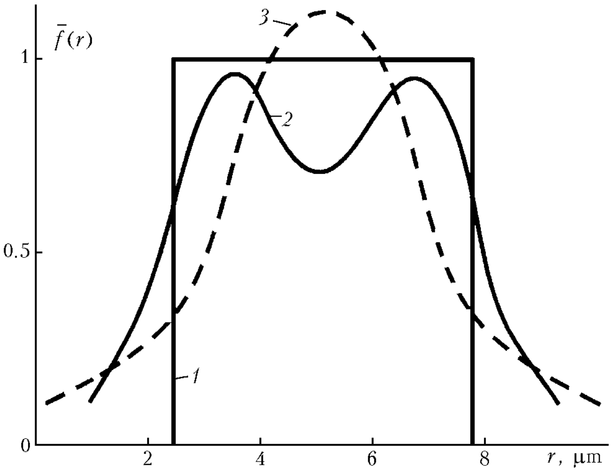

Fig. 1. Initial (*1*) and reconstructed bimodal (*2*) and unimodal (*3*) distributions.

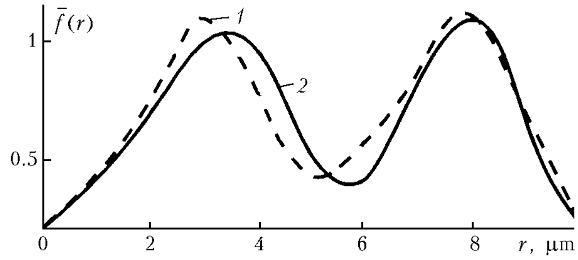

Fig. 2. Initial (*1*) and reconstructed (*2*) distributions.

Figure 1 presents the comparison of the initial distribution (stepwise function) with the first iteration (approximation) of the reconstructed dependence in the classes of unimodal and bimodal distributions. The depicted distributions well illustrate the ambiguity of distribution reconstruction from particular values of the scattering phase function. The ambiguity is excluded by involving some *a priori* information,[8–10] which is formulated here as restrictions imposed on the spectrum of output values for the generator of compositions.

In the considered situation, the main restriction is the preset number of different maxima in the generated determining function, which just determines the number of modes of the reconstructed distribution function. Emphasize that for the proposed solving technique the modality (qualitative form) of the reconstructed function is determined by involving *a priori* data, with which the quantitative dependence can be unambiguously drawn.

The effect of measurement errors on the result of reconstruction was modeled by replacing the true values of the scattering phase function $I(\Theta_j)$ with

$$I(\Theta_j) = [1 + pH_j]\, I(\Theta_j),$$

where H_j are random values distributed uniformly at the interval [–0.01; +0.01], p is the relative (in percent) error of measurement of the scattering phase function.

The results calculated at the first iteration (approximation) are shown in Fig. 2. They demonstrate the agreement between the initial and reconstructed bimodal distributions $f(r)$ with the error level p = 25% typical of measurements of the scattering phase function.

From the results shown in Fig. 2 we can see a rather close qualitative and quantitative agreement (the maximal discrepancy no higher than 11%) between the initial and reconstructed dependences even at the first iteration, that confirms the high computational capabilities of the proposed technique. The mean time for computation of a typical variant on an IBM PC (Pentium 200 MHz) was less than one minute.

2. Modified spectral transparency method

The method for determination of spherical particle mean sizes based on the fact that the averaged extinction efficiency factor $\overline{Q}$ is invariant with respect to the form of $f(r)$ was proposed in Ref. 1. The algorithm for finding the mean volume-surface radius r_{32} and the mass concentration of condensed particles C from the measured values of the optical thickness τ for two wavelengths of the sensing radiation is reduced to solving the system of equations

$$\tau_j = \frac{3C\,\ell\,\overline{Q}(\rho_{32,j})}{\rho_k\, r_{32}}, \quad j = 1, 2, \qquad (6)$$

where ρ_k is the density of the particle material; ℓ is the optical length of the path; $\rho_{32,j} = 2\pi r_{32}/\lambda_j$ is the average parameter of diffraction.

Determination of the parameters of polymodal distributions $f(r)$ by the spectral transparency method requires, as a rule, multifrequency sensing in a wide wavelength λ range. As is known, the method was applied to diagnostics of disperse media with the bimodal particle size distribution, and this range was decreased due to "fraction" interaction of radiation with polydisperse particles.[6]

Consider the possibility of using this method to estimate the parameters of bimodal distributions consisting of particles of two sharply different size fractions. In this case, $f(r)$ can be represented as a sum $f(r) = f_1(r) + f_2(r)$ with each $f_i(r)$ characterized by its own values of r_0 and r_{32}, C. Assume that $\overline{Q}$ is invariant regardless of the form of $f(r)$ within the framework of interaction by the Mie mechanism,[3,5] that is, $\overline{Q} = \overline{Q}\,(\rho_{32}, i)$. For diagnostics of such systems, the wavelengths are selected from the condition

$$\Lambda_{ij} \approx r_{0i}/\rho_{\rm m},$$

where $\Lambda_{ij} = \lambda_{ij}/\pi$ (i, j = 1, 2); $\rho_{\rm m}$ is the coordinate of the first maximum of the extinction efficiency factor $Q(\rho)$ for the particles under study.

The system of equations for r_{32} and C has the form

$$\begin{cases} \tau_{i1} = \dfrac{3C_i\,\ell}{\rho_k\, r_{32,i}}\overline{Q}(\Lambda_{i1}), \\ \tau_{i2} = \dfrac{3C_i\,\ell}{\rho_k\, r_{32,i}}\overline{Q}(\Lambda_{i2}), \end{cases} \quad i = 1, 2, \qquad (7)$$

where

$$\overline{Q}(\Lambda_{ij}) = \frac{\int_0^{\rho_*} Q(\rho)\,\rho^2 f_i(\rho\,\Lambda_{ij})\,d\rho}{\int_0^{\rho_*} \rho^2 f_i(\rho\,\Lambda_{ij})\,d\rho}, \qquad (8)$$

and ρ_* can be found from the equation

$$\int_0^{\rho_*} Q(\rho)\,(\rho - \rho_{\rm m})\,d\rho = 0. \qquad (9)$$

Possible realization of the method was studied by solving the model problem for $f(r)$ in the following form:

$$f(r) = b_1\, r^{\beta_1} \exp[-(r/r_{01})^2] + $$
$$+ b_2\, r^{\beta_2} \exp[-(r/r_{02})^2] \qquad (10)$$

(sum of generalized gamma distributions), as well as by comparing the results of optical diagnostics with the data of sampling (slide deposition) of aluminum oxide particles sprayed by a plasmotrone jet. The tentative results of these studies have shown that if the needed *a priori* information about $f(r)$ is available, it is possible to determine the parameters of the bimodal distributions of the considered class by the spectral transparency method with a limited number of sensing wavelengths.

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