Modified method of a spectral transmission for measuring the aerosol disperse composition

V.A. Arkhipov, I.R. Akhmadeev, S.S. Bondarchuk, B.I. Vorozhtsov, A.A. Pavlenko, and M.G. Potapov

Institute for Problems of Chemical and Energetic Technologies, Siberian Branch of the Russian Academy of Sciences, Biysk

Received February 14, 2006

We analyze a modified method of a spectral transmission and experimental setup for measuring aerosol-particle mean size in the optically dense high-temperature two-phase flows. The method is based on laser beam attenuation by a cloud of particles using sounding radiation of a limited number of wavelengths. Experimental results on the condensed aluminum particle size in the heterogeneous products of burning are presented.

Introduction

Contactless methods based on solving inverse problems of optics of aerosol, such as the method of scattering phase function at small scattering angles, the method of total scattering phase function, and the method of spectral transmission[1,2] are widely used in measuring the aerosol-particle size spectra. However, application of these methods to diagnostics of the high-temperature optically dense aerosol systems, in particular, heterogeneous plasma of the products of burning with high concentration of condensed particles, is difficult because of the presence of intense emission of the aerosol particles themselves.

In this paper we consider a modified method of spectral transmission based on measuring the spectral transmission coefficients of a cloud of particulate matter using sounding radiation of a limited number of wavelengths.[2,3] This method does not enable one to determine the particle size distribution function $f(a)$, however, it has some advantages in measuring the mean particle size, in particular, mean volume–surface diameter a_{32}, such as simplicity of the instrumental setup, its easy adjustment, possibility of making diagnostics of the high-temperature two-phase flows of high optical density.

Measurement technique

The idea of this method is in solving the inverse problem stated by the integral equation

$$\tau_\lambda = \frac{\pi C_n l}{4}\int_0^\infty a^2 Q(a,\lambda,m) f(a)\mathrm{d}a, \qquad (1)$$

where τ_λ is the spectral optical thickness, C_n is the number density of particles, λ is the wavelength of sounding radiation, Q is the extinction efficiency factor of an individual particle, l is the optical path length, a is the diameter of a particle, m is the complex refractive index of the particulate matter.

The averaged extinction efficiency factor is defined by the formula

$$\bar{Q}(a,\lambda,m) = \frac{\int_0^\infty Q(a,\lambda,m) a^2 f(a)\mathrm{d}a}{\int_0^\infty a^2 f(a)\mathrm{d}a}. \qquad (2)$$

Replacing the number density C_n by the mass concentration

$$C_m = C_n \frac{\pi\rho_p}{6}\int_0^\infty a^3 f(a)\mathrm{d}a, \qquad (3)$$

we obtain the formula for the optical thickness

$$\tau_\lambda = \frac{1.5 C_m l \bar{Q}(a,\lambda,m)}{\rho_p a_{32}}, \qquad (4)$$

where ρ_p is the density of the particulate matter;

$$a_{32} = \frac{\int_0^\infty a^3 f(a)\mathrm{d}a}{\int_0^\infty a^2 f(a)\mathrm{d}a}$$

is the mean volume–surface particle diameter.

Physical model of the method is based on the interaction of a monochromatic radiation with a polydisperse medium through the Mie mechanism and conservation of invariance of the mean extinction efficiency factor relative to the shape of the particle size distribution function. Validity of this assumption is caused by the fact that $\bar{Q}(\lambda,m)$ is determined by integrals of $f(a)$ and, hence, it is weakly sensitive to the peculiarities of the $f(a)$ behavior in the range of the particle size considered.[2,4]

Under certain conditions, the mean extinction efficiency factor does not depend on the shape of $f(a)$, while being a function of the mean volume–surface diameter of particles a_{32} at a set wavelength of sounding radiation. It is the most important optical property of the two-phase polydisperse media.

The problem on determination of the particle size by this method is reduced to measuring the spectral optical thickness of a disperse medium at two wavelengths λ_1 and λ_2 and calculating the mean extinction efficiency factors at the same wavelengths.

The ratio of experimentally measured optical thicknesses at two wavelengths is equal to the ratio of the mean extinction efficiency factors and is the function of the mean particle size:

$$\frac{\tau_{\lambda i}}{\tau_{\lambda j}} = \frac{\bar{Q}(a_{32}, \lambda_i, m)}{\bar{Q}(a_{32}, \lambda_j, m)} = F_{ij}(a_{32}). \quad (5)$$

The range of mean particle size measurable depends on the wavelengths of sounding radiation chosen. Thus, at $\lambda_i = 0.63$ μm and $\lambda_j = 3.39$ μm the range of a_{32} measurable is from 0.5 to 4 μm.

Three wavelengths of sounding radiation were used in the considered setup, namely, $\lambda_1 = 0.63$, $\lambda_2 = 1.15$, and $\lambda_3 = 3.39$ μm, and the ratios of experimentally measured optical thicknesses F_{21}, F_{31}, and F_{32} were determined.

The extinction efficiency factors of individual particles were calculated by the exact expressions of Mie theory using logarithmic derivatives of the Riccati–Bessel functions.[5] Optical constants of the aluminum oxide particles were taken from Ref. 6. The mean extinction efficiency factors $\bar{Q}(a_{32}, \lambda_i, m)$ are shown in Fig. 1 as functions of the mean volume–surface diameter a_{32}.

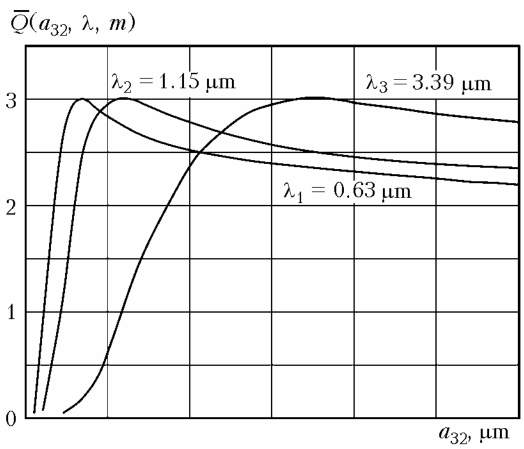

Fig. 1.

Thus, one can determine a_{32} from the experimentally measured $\tau_{\lambda i}$ and calculated values $F_{ij}(a_{32})$. The number density of particles in the measurement zone was determined by the following formula

$$C_{\rm m} = \frac{\tau_\lambda \rho_{\rm p} a_{32}}{1.5 l \bar{Q}(a_{32}, \lambda, m)} \quad (6)$$

assuming that the optical path length to be *a priori* known or determined experimentally.

In practical realization of the multiwavelength sensing of two-phase media, the choice of the radiation wavelengths is necessary, the measurements at which bear information on the particle size spectrum.

The main condition for choosing λ_i in realizing this method is the well pronounced dependence of the mean extinction efficiency factors on the mean volume–surface diameter of particles. The position of the maximum of the functions $\bar{Q}(\rho_{32})$ is well described by the formula[4]:

$$\rho_{32}^{\rm o} = \frac{\pi}{\lambda} a_{32}^{\rm o} = 1 + \frac{4}{|m|^2 - 1}, \quad (7)$$

where the superscript "o" determines the values of the mean diffraction parameter ρ_{32} and the particle size a_{32}, at which the function $\bar{Q}(\rho_{32})$ has an extreme.

It follows from the condition (7) that setting *a priori* the range of the measurable mean size of particles under study ($a_{32}^{\min}$, $a_{32}^{\max}$), determines the radiation wavelengths to be near the following values[7]:

$$\lambda^{\min} = \pi a_{32}^{\min} \left(\frac{|m|^2 - 1}{|m|^2 + 3} \right), \quad (8)$$

$$\lambda^{\max} = \pi a_{32}^{\max} \left(\frac{|m|^2 - 1}{|m|^2 + 3} \right). \quad (9)$$

At $\lambda_i < \lambda^{\min}$, the mean size of fine particles of the medium under study will be measured in the experiment, while at $\lambda_i > \lambda^{\max}$ of the coarse particles.

The values of the ratios of the mean extinction efficiency factors $F(a_{32})$ at three wavelengths of sounding radiation calculated by Eq. (5) are presented in Table 1.

Table 1. Ratios of the mean extinction efficiency factors ($\lambda_1 = 0.63$, $\lambda_2 = 1.15$, and $\lambda_3 = 3.39$ μm)

a_{32}, μm	F_{21}	F_{31}	F_{32}	a_{32}, μm	F_{21}	F_{31}	F_{32}
1.5	1.117	0.648	0.580	5.0	1.065	1.281	1.202
2.0	1.116	0.946	0.847	5.5	1.089	1.303	1.196
2.5	1.100	1.139	1.031	6.0	1.063	1.254	1.180
3.0	1.125	1.270	1.128	6.5	1.067	1.231	1.152
3.5	1.087	1.308	1.203	7.0	1.099	1.226	1.114
4.0	1.064	1.296	1.218	7.5	1.056	1.222	1.157
4.5	1.089	1.310	1.203	8.0	1.050	1.211	1.151

The maximum measured particle size a_{32} in this case is determined by the value, at which the corresponding function $F(a_{32})$ has its maximum.

The ranges ($a_{32}^{\min}$, $a_{32}^{\max}$) of measured a_{32} values were determined based on calculated dependences $\bar{Q}(a_{32})$ and $F(a_{32})$ for the corresponding pairs of wavelengths (Table 2).

Table 2. Ranges of the mean particle size

λ_1, μm	λ_2, μm	$a_{32}^{\min}$, μm	$a_{32}^{\max}$, μm
0.63	1.15	0	1.5
0.63	3.39	0	3.5
1.15	3.39	0.1	3.6

As was mentioned above, the method is based on the Mie theory to describe the interaction of radiation with a particle within the frameworks of single scattering of light by independent spherical homogeneous particles. As a rule, the condensed particles of metal oxides in high-temperature two-phase plasma flows of the products of burning are liquid droplets. A droplet under the effect of surface tension takes the shape close to spherical (at Weber numbers less or equal to 1–5).[8]

The contribution of multiple scattering becomes essential at the values of the optical thickness τ > 10–18.[9] In that case deviations from the Bouguer law occur, and the method of spectral transmission can result in large errors in determining the aerosol particle size.[10]

Besides, serious problems appear in measuring the spectral transmission coefficients of optically dense flows related to isolation of the sounding radiation against the background of the emission of the plasma itself. Thus, real applicability limits of the method are for optical thickness that does not exceed the values τ_λ = 3–4.[2]

Experimental setup

The study of the disperse parameters of aerosol flows in high-temperature products of burning in the frameworks of the considered method requires development of corresponding instrumentation that would meet quite strict requirements. Operation of such instrumentation complexes is based on the principle of measuring extinction of laser radiation passed trough the flow.

The laser measurement complex consists of a block of emitters, a block of photodetectors, and a recording device. A number of requirements are imposed on all components of the complex: wide range of intensities and wavelengths of the laser radiation, high spectral sensitivity and linear dependence of the output signals of the photodetectors, wide dynamical range of the detectable signals and frequency stability of the recording devices.[3]

All these conditions were realized in a specially designed three-wavelength He–Ne radiation source (Fig. 2), which consists of a commercial LG-75 laser (the output power of 25 mW) and the optical–mechanical block including the spherical mirror covered with aluminum (with the focal length of 2 m) and a modulator with a perforated disc.[11]

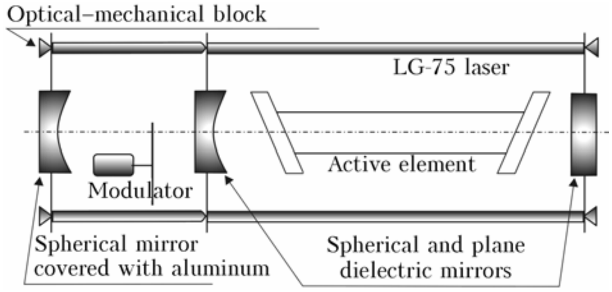

Fig. 2. Functional block-diagram of the three-wavelength radiation source.

The optical–mechanical block is an adjustment device from a commercial laser attached to the end of the LG-75 laser by means of small invar rods thus comprising the single unit with it. Resonator of the LG-75 laser for radiation at the wavelength λ = 0.63 μm is placed inside the resonator for λ = 3.39 and 1.15 μm. The output mirror is common for all wavelengths. Modulator opens and closes the mirror covered with aluminum from the side of the active element by means of a perforated disc, then the external resonator for λ = 3.39 and 1.15 μm between dielectric mirrors is switched on and off.

When the external resonator has been switched on, the conditions are provided for generation at λ = 3.39 μm. Simultaneously occurs the generation at the wavelength λ = 1.15 μm. When modulator closes the mirror, the conditions are provided for generation at λ = 0.63 μm in resonator formed by dielectric mirrors of LG-75 laser. As a result, the modulated laser radiation appears at three wavelengths directed along the same optical path.

The photodetection block with the system of wavelength selection was designed for recording laser radiation at three wavelengths (Fig. 3). The laser beam in the photodetection block is divided into three beams by means of the system of dividing semi-transparent optical plates with a dielectric coating. Radiation at the wavelengths of 0.63 and 1.15 μm is recorded with FD-24K photodiodes. The interference filter is used for selection of the wavelength of 0.63 μm, and the silicon filter is used for λ = 1.15 μm. Radiation at the wavelength of 3.39 μm is recorded by means of an MG-30 pyroelectric detector with a germanium filter.

Calibration of the measurement path was carried out in order to control the linearity of the photodetection and identification of the optical thickness of the two-phase flow by use of standard values of the calibration optical filters. The block-diagram of the measurement setup is shown in Fig. 4.

Fig. 3. External view of the three-wavelength laser with photodetection block.

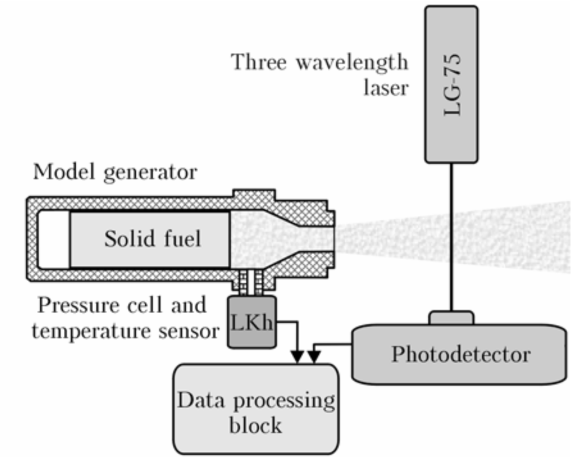

Fig. 4. Block-diagram of measurements of the parameters of the products of burning.

Fig. 5.

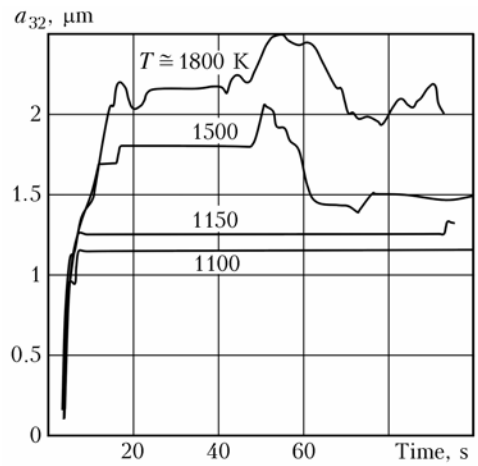

Fig. 6.

Measurement results

The technique was tested and validated by analyzing the condensed products of burning (aluminum oxide) of the model compounds of mixed solid fuels based on ammonium perchlorate, butyl rubber, and 10% aluminum powder of the ASD-4 type. Cylinder samples of solid fuels of 47-mm diameter and 81-mm length having mass of 0.25 kg were used. Measurements were carried out in a heterogeneous flow of the products of burning in a nozzleless gas generator with the outlet diameter of 10.8 to 13.3 mm. The values of the spectral optical thickness τ_λ measured in the experiments did not exceed 1 to 3. The external view of the flow of the products of burning during the operation of the model generator is shown in Fig. 5.

Typical experimental data of the obtained dependences of the particle size a_{32} on time are shown in Fig. 6 for different mean values of temperatures realized in the burning chamber.

Non-monotonic view of the dependences at high temperatures is caused by quite strong nonstationary behavior of the inner ballistic parameters realized under these conditions, in particular, by moving the boundary of the critical cross section and the change of the length of the zone of high speed of the flow.

Conclusion

Thus, the proposed modified method of spectral transmission for measuring the aerosol disperse composition is characterized by a sufficient simplicity of instrumentation and its alignment, as well as by the possibility of performing diagnostics of two-phase flows of large optical density.

The instrumentation complex developed and the software for determination of the parameters of the products of burning were tested by means of analysis of the disperse composition of the products of burning of model mixed solid fuels at burning in a nozzleless generator.

Acknowledgments

The work was supported in part by the Russian Foundation for Basic Research (grants Nos. 05-03-32729 and 05-08-18237) and ISTC project No. 3305.

References

1. V.E. Zuev, B.V. Kaul, I.V. Samokhvalov, K.I. Kirkov, and V.I. Tsanev, *Laser Sensing of Industrial Aerosols* (Nauka, Novosibirsk, 1986), 188 pp.
2. V.A. Arkhipov, *Laser Methods for Diagnostics of Heterogeneous Flows.* Manual (Tomsk State University, Tomsk, 1987), 140 pp.
3. B.I. Vorozhtsov, M.G. Potapov, A.A. Pavlenko, V.P. Lushev, Yu.A. Galenko, and Yu.V. Khrustalev, Atmos. Oceanic Opt. **10**, No. 7, 518–520 (1997).
4. A.P. Prishivalko and E.K. Naumenko, *"Light scattering by spherical particles and polydisperse systems."* Preprint No. 59, Institute of Physics, Academy of Sciences of Belarus, Minsk (1972), 61 pp.
5. D. Deirmendjian, *Electromagnetic Waves Scattering on Spherical Polydispersions* (Elsevier, New York, 1969).
6. D.A. Gryvnak and D.E. Burch, J. Opt. Soc. Am. **55**, No. 6, 625–629 (1965).
7. A. Pavlenko, V. Arkhipov, B. Vorozhtsov, I. Akhmadeev, and M. Potapov, in: *Abstracts of XII Joint Int. Symp. on Atmospheric and Oceanic Optics. Atmospheric Physics,* IAO SB RAS, Tomsk (2005), p. 148.
8. V.A. Arkhipov, I.M. Vasenin, V.G. Butov, A.A. Glazunov, and V.F. Trofimov, *Gas Dynamics of Two-Phase Flows in Nozzles* (Tomsk State University, Tomsk, 1986), 262 pp.
9. B.A. Saveliev. Izv. Vyssh. Uchebn. Zaved. SSSR, Ser. Fizika, No. 12, 136–138 (1967).
10. V.E. Zuev and M.V. Kabanov, *Transfer of Optical Signals in the Earth's Atmosphere* (Sov. Radio, Moscow, 1977), 368 pp.
11. V.P. Lushev, O.A. Moskalets, A.A. Pavlenko, and E.V. Podlesnaya, Prib. Tekh. Eksp., No. 2, 178–179 (1990).