Calculation method for particle mean diameter and particle size distribution function under dependent model algorithm

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In total light scattering particle sizing technique, the relationship among Sauter mean diameter D_{32}, mean extinction efficiency $\overline{Q}$, and particle size distribution function is studied in order to inverse the mean diameter and particle size distribution simply. We propose a method which utilizes the mean extinction efficiency ratio at only two selected wavelengths to solve D_{32} and then to inverse the particle size distribution associated with $\overline{Q}$ and D_{32}. Numerical simulation results show that the particle size distribution is inversed accurately with this method, and the number of wavelengths used is reduced to the greatest extent in the measurement range. The calculation method has the advantages of simplicity and rapidness.

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Light scattering particle sizing techniques have been widely used during the recent years since they provide an important tool for the characterization of a large number of industrial production processes. These techniques mostly contain the total light scattering, angle light scattering, diffraction light scattering, and dynamic light scattering, and the measurement covers the range from nanometers to millimeters. Among these techniques, the total light scattering technique is probably the most attractive one which does not need absolute calibration. It can in fact be used for in-line monitoring of micron or sub-micron particle systems, thus providing real time measurements of both particle size distribution and particle concentration. The total light scattering technique is very simple in terms of measurement principle and very convenient with regard to the optical arrangement. With the urgent demand for *in situ* particle sizing, the total light scattering technique has grown rapidly. In total light scattering particle sizing technique, the upper limit of measurement range of particle mean diameter cannot exceed 1—2 μm with the conventional two-wavelength method or multiple two-wavelength method generally, otherwise the inversion results of particle size distribution may be multivalued. In this paper, we propose a method which utilizes the mean extinction efficiency ratio at only two selected wavelengths to inverse the particle mean diameter and particle size distribution function under the dependent model algorithm simply.

The total light scattering technique is based on the light scattering theory. When a beam of parallel monochromatic radiation light passes through a suspension of particle system with a refraction index different from that of the dispersant medium, scattering and absorbing lead to an attenuation of the transmitted light. According to the Lambert-Beer law, if the suspension of particle system is polydisperse spheres and the multiple scattering and interaction effects can be neglected, the transmitted light intensity I is defined as[1]

$$\ln\frac{I}{I_0} = -\frac{\pi}{4} \times L \times \int_{D_{\min}}^{D_{\max}} Q(\lambda, m, D) \times N(D) \times D^2 \mathrm{d}D, \quad (1)$$

where I_0 is the incident light intensity (i.e. the intensity of the transmitted light through the suspending medium in the absence of suspended particles), the extinction value I/I_0 is provided by actual measurement; L is the thickness of the particle system; $Q(\lambda, m, D)$ is the Mie extinction efficiency which is a complex function of particle diameter D, wavelength λ in the medium, and relative refractive index m (the ratio between the particle and medium refractive index), and can be calculated from Mie theory[2,3]; $N(D)$ is the number concentration of particle system (in unit of cm^{-3}) which is the particle size distribution function to be measured.

Equation (1) is Fredholm integral equation of the first kind which is ill conditioned[4]. The particle size distribution function is inversed by some solution algorithms based on the measurement of turbidity at multi-wavelength in total light scattering technique. The developments of stable algorithms to inverse the particle size distribution function have long been a subject of research effort. These algorithms can be divided into two categories[5]. The first category is independent model algorithm in which no *a prior* information about the particle size distribution is available and the particle size distribution function is inversed by the discrete liner equation set. The second one is dependent model algorithm in which some *a prior* information is available and the true particle size distribution function is inversed using a certain optimization algorithm. Actually, many particle systems to be measured often conform to some two-parameter size distributions, so using the dependent model algorithm to inverse the particle size distribution function is simpler. Different types of particle size distribution functions have been used in dependent model algorithm studies such as the Rosin-Rammler (R-R) function, the log-normal function, the gamma function, and so on. Among them, the most widely used one is the R-R distribution function.

The R-R distribution function is used here as an example to introduce the calculation method. The mathematical representation of R-R particle size distribution function can be defined as[1]

$$f(D) = \frac{k}{\overline{D}} \times (\frac{D}{\overline{D}})^{k-1} \times \exp(-(\frac{D}{\overline{D}})^k), \quad (2)$$

where D is the particle diameter (in unit of μm), $\overline{D}$ is the size parameter (in unit of μm) which denotes the particle volume with diameter smaller than D accounting for 63.21% of the total particle volume, k is the dimensionless distribution parameter.

The relationship between $N(D)$ and $f(D)$ is written as

$$f(D)\mathrm{d}D = \frac{\pi}{6}D^3 N(D)\mathrm{d}D. \quad (3)$$

In total light scattering particle sizing technique, the mean extinction efficiency $\overline{Q}$ and Sauter mean diameter D_{32} are defined in terms of the principle of equivalent extinction efficiency as

$$\overline{Q} = \frac{\int_{D_{\min}}^{D_{\max}} Q(\lambda, m, D) \times N(D) \times D^2 \mathrm{d}D}{\int_{D_{\min}}^{D_{\max}} N(D) \times D^2 \mathrm{d}D}, \quad (4)$$

$$D_{32} = \frac{\int_{D_{\min}}^{D_{\max}} N(D) \times D^3 \mathrm{d}D}{\int_{D_{\min}}^{D_{\max}} N(D) \times D^2 \mathrm{d}D}. \quad (5)$$

Thus the polydisperse particle system may be converted into a monodisperse particle system.

The transmitted light intensity I can be rewritten as

$$\ln\frac{I}{I_0} = -\frac{\pi}{4} LND_{32}^2 \overline{Q}(\lambda, m, D_{32}). \quad (6)$$

When the diameter D of particle system varies from zero to positive infinity, D_{32} can be expressed as

$$D_{32} = \frac{\int_0^{+\infty} N(D) \times D^3 \mathrm{d}D}{\int_0^{+\infty} N(D) \times D^2 \mathrm{d}D} = \frac{\overline{D}}{\chi(1 - \frac{1}{k})}, \quad (7)$$

where χ is the gamma function, $\chi(\alpha) = \int_0^\infty \chi^{\alpha-1} \mathrm{e}^{-\chi} \mathrm{d}\chi$.

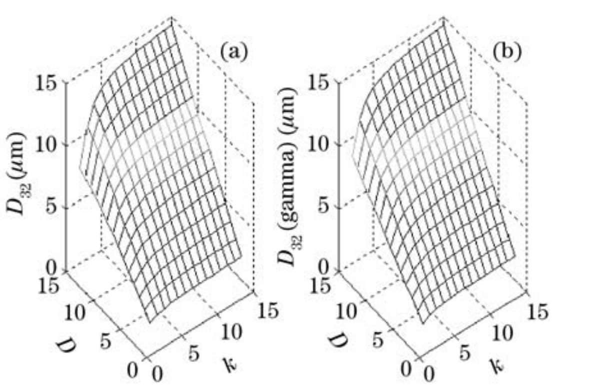

Fig. 1. Relationship between D_{32} and R-R distribution parameters k and $\overline{D}$ calculated by (a) Eq. (5) and (b) Eq. (7), respectively.

Figure 1 shows D_{32} calculated by Eqs. (5) and (7) respectively as a function of distribution parameters $\overline{D}$ and k within the measurement range of D from 0 to 20 μm, which indicates that the calculated deviations of D_{32} between the given interval and the infinite interval are so small that it is simple to use Eq. (7) as the calculation formula for D_{32} in the given interval.

D_{32} can be determined by measuring the extinction values I/I_0 at the two selected wavelengths (two-wavelength method). The calculation formulas are

$$\ln\frac{I_1}{I_{01}} = -\frac{\pi}{4} LND_{32}^2 \overline{Q}(\lambda_1, m, D_{32}), \quad (8)$$

$$\ln\frac{I_2}{I_{02}} = -\frac{\pi}{4} LND_{32}^2 \overline{Q}(\lambda_2, m, D_{32}). \quad (9)$$

Since the twice measurements are within the same particle system and at the same time, L, N, and D_{32} in Eqs. (8) and (9) are completely uniform, the extinction ratio R_{12} is given by

$$R_{12} = \frac{\ln\frac{I_1}{I_{01}}}{\ln\frac{I_2}{I_{02}}} = \frac{\overline{Q}(\lambda_1, D_{32})}{\overline{Q}(\lambda_2, D_{32})}. \quad (10)$$

In Eq. (10), the extinction ratio R_{12} can be obtained from actual measurements of extinction values, and $\overline{Q}$ is the unary function of D_{32} (with known values of m and λ). Therefore the extinction ratio R_{12} at the two wavelengths is only related to D_{32}[6].

Figure 2 shows $\overline{Q}$ as a function of D_{32} for different R-R distribution at $\lambda_1 = 0.4$ μm and $\lambda_2 = 10.6$ μm, and the distribution parameter k is chosen in a relatively wide range from 3.5 to 8.5. As can be seen from the figure, when $\lambda_1 = 0.4$ μm, $D_{32} > 2$ μm, $\overline{Q}$ is not very much dependent on the particle distribution parameter $\overline{D}$ and

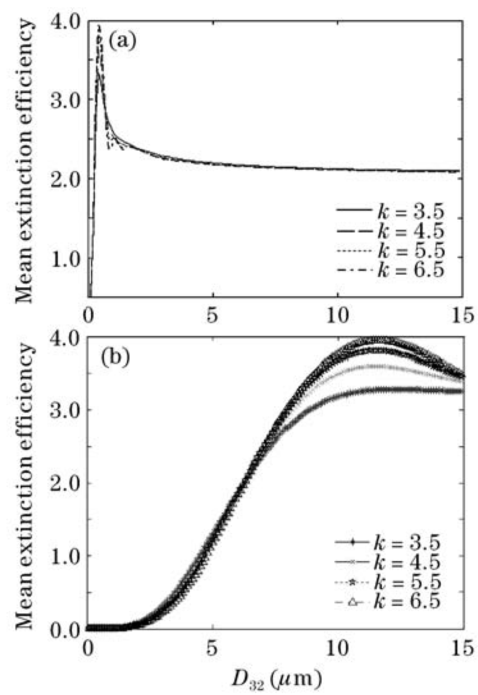

Fig. 2. Mean extinction efficiency for different R-R distribution functions. (a) $\lambda = 0.4$ μm; (b) $\lambda = 10.6$ μm.

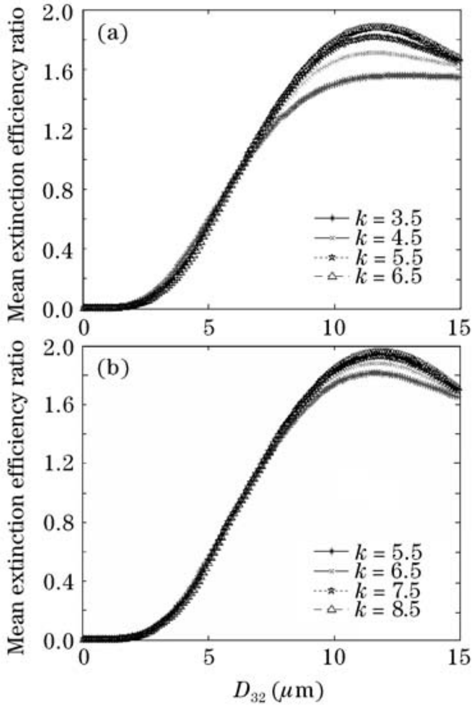

Fig. 3. Mean extinction efficiency ratio at $\lambda_1 = 0.4$ μm and $\lambda_2 = 10.6$ μm. (a) $k = 3.5 - 5.5$; (b) $k = 5.5 - 8.5$.

k, that is, the shape of the particle distribution function has little effect on the curves of $\overline{Q}$; when $\lambda_2 = 10.6$ μm, $D_{32} < 9$ μm, the similar conclusion can also be drawn. Therefore when 2 μm $< D_{32} <$ 9 μm, the shape of particle distribution function has little effect on R_{12} at $\lambda_1 = 0.4$ μm and $\lambda_2 = 10.6$ μm. Most importantly, R_{12} varies monotonously with D_{32} within the measurement range. Figure 3 proves the conclusions, Fig. 3(b) indicates that the measurable range of D_{32} for the case of $5.5 < k < 8.5$ is larger than that for the case of $3.5 < k < 5.5$.

From Fig. 3, we can come to a conclusion that utilizing the extinction efficiency ratio R_{12} at the two selected wavelengths can determine Sauter mean diameter D_{32} uniquely without knowing the particle size distribution when D_{32} is in the measurement range from 2 to 9 μm. This is also applicable for bi-peak and multi-peak particle size distributions. Having solved D_{32} from the extinction efficiency ratio R_{12} at the two selected wavelengths, one may utilize the mean extinction efficiency curves known in advance to determine $\overline{Q}$ correspondingly. The measurement range of D_{32} is 2—9 μm, and that of particle diameter D is from 0.1 to 10 μm which is the optimal measurement range in total light scattering technique.

When $D < 10$ μm, the variation of $Q(\lambda, m, D)$ with particle diameter D at $\lambda_2 = 10.6$ μm may be seen as monotonic[7], furthermore the variation of $\overline{Q}$ with D_{32} may also be seen as monotonic in the measurement range of D_{32} of 2—9 μm at $\lambda_2 = 10.6$ μm (see Fig. 2). So the particle distribution parameter $\overline{D}$ and k can be determined entirely by solving Eqs. (4) and (7), and they are the unique definite solution.

Table 1 gives a part of the numerical simulation calculation results of particle distribution parameter $\overline{D}$ and k which are inversed from the known D_{32} and $\overline{Q}$. The results show that the deviations of $\overline{D}$ and k between the set value and calculated value are very small in the measurement range.

Table 1. A Part of Numerical Simulation Results

D_{32} (μm)	$\overline{Q}$	k Set	k Calculated	$\overline{D}$ (μm) Set	$\overline{D}$ (μm) Calculated
2.5	0.1659	3.5	3.4976	3.1903	3.1907
6.8	2.2731	3.5	3.5262	8.6826	8.6551
2.6	0.1575	4.5	4.4966	3.0947	3.0949
7.9	2.8682	4.5	4.5058	9.4038	9.3995
3.2	0.2961	5.5	5.4963	3.6625	3.6625
8.0	2.9862	5.5	5.5017	9.1552	9.1548
3.9	0.5394	6.5	6.4921	4.3533	4.3538
8.2	3.1279	6.5	6.5014	9.1527	9.1523

When measuring the extinction value I/I_0, if the measurement error is very large, the mean extinction efficiency $\overline{Q}$ curves will be changed, and it is difficult to determine D_{32} and $\overline{Q}$ accurately. So it is necessary to ensure the stabilization of light source. Using laser to generate the two selected wavelengths can improve the luminance and collimation of light source.

A novel calculation method for inversing the particle mean diameter and particle size distribution function has been presented. This method has overcome the difficulties of the very small upper limit of the mean diameter measurement range and the multivalued inversion results of particle size distribution using the conventional two-wavelength method. Using the ratio at two wavelengths may reduce the effects of the fluctuation of light source, the interference of external stray light, and the effect of pollution in the observation windows on the measurement results[8]. Numerical simulation results show that the particle size distribution function is inversed accurately with this method in the measurement range of D_{32} from 2 to 9 μm. The calculation method is simple and fit for the in-line measurement of particle size and concentration.

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