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Dependence of the forward light scattering on the refractive index of particles

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ABSTRACT

In particle sizing technique based on forward light scattering, the scattered light signal (SLS) is closely related to the relative refractive index (RRI) of the particles to the surrounding, especially when the particles are transparent (or weakly absorbent) and the particles are small in size. The interference between the diffraction (Diff) and the multiple internal reflections (MIR) of scattered light can lead to the oscillation of the SLS on RRI and the abnormal intervals, especially for narrowly-distributed small particle systems. This makes the inverse problem more difficult. In order to improve the inverse results, Tikhonov regularization algorithm with B-spline functions is proposed, in which the matrix element is calculated for a range of particle sizes instead using the mean particle diameter of size fractions. In this way, the influence of abnormal intervals on the inverse results can be eliminated. In addition, for measurements on narrowly distributed small particles, it is suggested to detect the SLS in a wider scattering angle to include more information.

1. Introduction

Particle size measurement is one of the most important and fundamental measurements in particle analysis. Light scattering methods have been widely used because of its advantages of wide measurable particle size range, good reproducibility, high precision and being non-invasive [1–3]. Forward light scattering (FLS) method uses the relationship between the scattered light signals in the range of small scattering angles and the particle size to get the information of particle size distribution (PSD). In the earlier FLS method, the scattered signal is described by the Fraunhofer diffraction (FD) theory which establishes the reciprocal dependence of the particle size on the diffraction angle approximately. The FD theory can be used only when the particle size is much larger than the wavelength of incident light and the relative refractive index (RRI) of the particles to the surrounding medium is far away from 1. Once the particle size is small, the distribution of the scattered light will be drastically different from that of the diffracted light. In terms of the transparent particle, of whom the RRI is close to 1, the transmitted light will interfere with the diffraction, producing anomalous diffraction. This kind of light intensity distribution cannot be described by the FD theory even for large particles [4]. Therefore, the FD theory is replaced by the classical Mie theory. The use of Mie theory extends the low limit of the measurable particle size of the FLS method. However, the inverse calculation requires the information of the refractive index of particles and the consistency between the input refractive index and the particle's refractive index becomes a prominent problem to the measurement results [5–7].

Much work concerning on the comparison between the FD theory and the Mie theory has been published during the past decades [8,9]. Recently, the effects on the measurement results in the FLS method caused by the mismatch between the input value of the refractive index and the real one of the sample are studied based on the Debye series expansion (DSE) in which the scattered light is taken as an interference of the diffraction (Diff), the surface reflection (SR) and the multiple internal reflections (MIR) [10,11]. It is found that, the scattered light signal (SLS) oscillates periodically along with the variation of the RRI, due to the interference between the Diff and the MIR light of the scattered light. This deteriorates the reciprocal dependence of the particle size on the scattering angle and hence leads to the abnormal intervals [12,13], which may lead to difficulties in the inverse procedure for extracting the particle size information. Up to now, such work is limited to the mono-dispersed or extremely narrowly-distributed particle systems. It is expected that, for widely-distributed particle systems, the abnormal intervals may be cancelled by the overlapping

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of the signals from particles of different sizes. Therefore, in this paper, the effects of particle size distribution on the abnormal intervals are studied with numerical simulations and experiments. Besides, the B-Spline functions are introduced to discretize the first kind Fredholm equations to eliminate the influence on the inverse results from the abnormal intervals.

2. Particle analysis based on forward light scattering (FLS)

The optical alignment of the FLS is schematically shown in Fig. 1. Particles are randomly dispersed in the sampling cell and are illuminated by a collimated beam. The photo-detector is located in the focal plane of the receiving lens and contains several ring-shaped units. The inner and outer radii of detecting units are distributed uniformly on the logarithmic scale. The receiving lens transforms the scattered light onto the photo-detector. The transmitted light passes through the hole at the center of the detector and is received by a transmission detector.

The SLS of a mono-disperse particle system on each detecting unit is given as:

$$e_{sca,i} = C \cdot \int_{\theta_{i,1}}^{\theta_{i,2}} i_{sca}(m,\alpha,\theta)\sin\theta d\theta \quad (i = 1,2,3,\ldots,M) \tag{1}$$

where $\theta_{i,1}$ and $\theta_{i,2}$ respectively represent the minimal and maximal angles of the ith detecting unit which are determined by the radial dimensions of detector and the focal length of the receiving lens. The constant C is decided by the transfer efficiency of the light signal and will be taken as 1. α is the dimensionless particle size parameter defined as $\alpha = \pi d/\lambda_0$ (d is the particle diameter and λ_0 is the wavelength of the incident light). m is the RRI defined as $m = m_P/m_s$ (m_P is the particle's refractive index and m_s is that of the surrounding medium). $i_{sca}(m,\alpha,\theta)$ is the scattered light intensity (SLI), which can be calculated with the classical Mie theory. M is the number of the detector units.

For a poly-dispersed particle system, the SLS that is received by the ith unit of the detector is a linear combination of those contributions from particles of different sizes [2,14].

$$E_{sca,i} = \int_{d_{min}}^{d_{max}} \left\{ \int_{\theta_{i,1}}^{\theta_{i,2}} i_{sca}(m,\alpha,\theta)\sin\theta \mathrm{d}\theta \right\} q_k(d) d^{-k} \mathrm{d}d \quad (i = 1,2,3,\ldots,M) \tag{2}$$

where d_{min} and d_{max} are the minimal and maximal diameters of the particles to be measured. The frequency function of the PSD $q_k(d)$ works as a weighting factor. $q_k(d)$ is the PSD in area when $k = 2$ and is the PSD in volume when $k = 3$. Eq. (2) is the first kind Fredholm integral and is traditionally discretized into the form of linear equations:

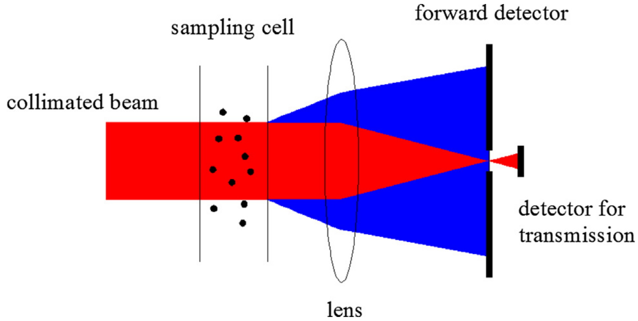

Fig. 1. Optical configuration with a collimated beam illumination.

$$\sum_{j=1}^{N} \underbrace{\pi \bar{d}_j^{-k} \int_{\theta_{i,1}}^{\theta_{i,2}} i_{sca}(m,\bar{\alpha}_j,\theta)\sin\theta \mathrm{d}\theta}_{A_{ij}} \underbrace{q_k(\bar{d}_j)\Delta d_j}_{Q_j} = E_i \tag{3}$$

which can also be expressed as a matrix equation:

$$\mathbf{AQ} = \mathbf{E} \tag{4}$$

Here, N is the number of particle size fractions. $\bar{d}_j$ and $\bar{\alpha}_j$ are the mean particle diameter and the corresponding dimensionless size parameter of the jth size fraction respectively. Δd_j is the width of the fraction. $\mathbf{Q} = (\mathbf{q}_1, \mathbf{q}_2, \ldots \mathbf{q_N})^{\mathbf{T}}$ is a vector representing the discretized PSD. Vector $\mathbf{E} = (\mathbf{E}_1, \mathbf{E}_2, \ldots, \mathbf{E}_N)^{\mathbf{T}}$ is the measured SLS. $\mathbf{A}$ is a $M \times N$ matrix which can be calculated theoretically with the Mie theory. The inverse problem for extracting the PSD $\mathbf{Q}$ from the matrix equation $\mathbf{AQ} = \mathbf{E}$ is a crucial work for particle size analysis, because the matrix equation is ill-conditioned. The inverse results depend on both the measurement and the inverse algorithm.

3. Dependence of the SLS on the RRI

3.1. Mono-dispersed particles

Firstly, we discuss the dependence of the SLS on the RRI for the mono-dispersed particle system. In order to exclude the effects from the width of the detecting unit on scattered signal, we assume that every detecting unit has the same radial width. Therefore, in the range of small scattering angles, the SLS within a unit radial width on the detector is given as:

$$\frac{d}{d\theta} e_{sca}(\theta) = i_{sca}(m,\alpha,\theta)\sin\theta \tag{5}$$

The SLSs calculated with the Mie theory are shown in Fig. 2, wherein the particle size parameter is $\alpha = 20$ and the RRIs are $m = 1.10$, $m = 1.50$ and $m = 1.90$ respectively. Numerical result of the FD approximation is also plotted for a comparison. It can be seen that, the profile of the SLS depends apparently on the RRI. Compared with the FD curve, the primary peak (labeled with P1) shifts inwards or outwards. The 2nd peak (labeled with P2) and the valley (labeled with V1) show the similar dependence on the RRI. Meanwhile, the magnitude of the 2nd peak P2 changes violently. This means the RRI affects the width of distribution of the SLS and also the relative heights of the primary and the 2nd peaks.

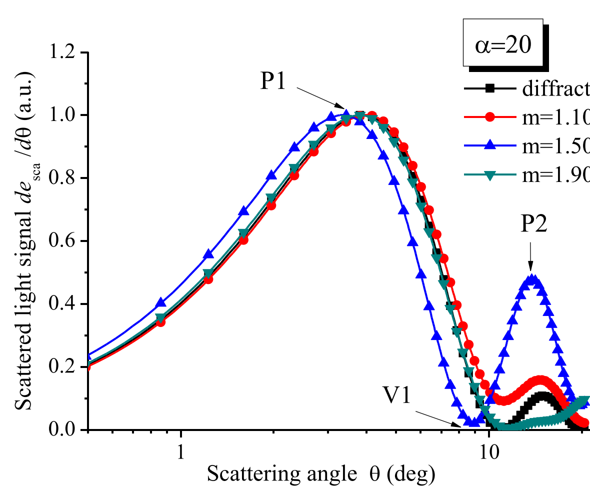

Fig. 2. Angular-dependent scattered light signals for $\alpha = 20$.