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<td><strong>Citation</strong></td>
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Influence of the number of detectors by laser scattering method for estimation of particle size

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Effect of the number of detectors on an inversion problem of a scattering pattern by laser scattering method based on Mie scattering model has been investigated. The influence of the number of detectors is obtained by comparing a given size distribution and a calculated size distribution by computer simulation and experimental method. An observing range of scattering angles is from 0.0007 to 2.5 rad. A non-linear iteration method is used for calculating particle size distribution. The number of detectors is changed from 6 to 81 elements by the computer simulation. The algorithm of the inversion problem is applied with mean diameters of log-normal distribution in a range from 0.546 to 214 μm at standard deviation of 0.27 and 0.68. Experimental results of certified mono-disperse polystyrene latex standards and a poly-disperse aluminum sample are obtained with 21, 41, and 81 elements detector, respectively. All tests are performed under conditions at diluted aqueous suspensions. Narrow size distribution is influenced by the number of detectors compared with wide size distribution. Not the number of physical detectors but the number of useful detectors affects the algorithm of the inversion problem. When the detector elements are over 20, the influence of the number of detectors is decreased. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4709493]

I. INTRODUCTION

A laser scattering (LS) method is one of the most useful techniques for measuring particle size distribution (PSD) due to wide dynamic range of measurement, good repeatability, and easy operation. The LS method is calculated PSD from angular dependence of light scattering intensity based on Mie scattering model. Commonly, the method of calculation from the light scattering is called an inversion problem. In order to solve the inversion problem, various methods such as constrained linear inversion method¹–⁵ and nonlinear iteration method¹, ⁶–¹⁴ can be considered. As the number of detectors is increased, the measured scattering pattern is accurate. Therefore, the number of detectors is related to the accuracy of calculated size distributions in the inversion method. The number of detectors that utilized in a commercial instrument is increased to as many as 126 elements recently.¹³ Each detector for the LS method has a response in a measurement range of particle size. If the number of detectors is changed, the response of the detector is also changed. We can expect that the optimum number of detectors is present for the LS method. However, the studies of the LS method have been used the fixed number of detectors previously. As the number of detectors is increased, the calculated PSD is thought to be accurate. However, any quantitative study of the effect of the number of detectors has not been investigated.

Authors report the relationship between the number of detectors and the accuracy of PSD for the LS method for the first time. A modified nonlinear iteration (MNLI) method based on Twomey¹⁴ is used for an algorithm of the inversion problem. The effect of the number of detectors using the LS method is studied through computer simulation and experimental method with mono-disperse and poly-disperse samples.

II. PRINCIPLE OF THE LS METHOD

When a globular particle is irradiated by a polarizing light, a scattering intensity at an angle is given from Mie scattering model.¹⁵–¹⁸ The scattered light intensity pattern from a large-diameter particle more than 50 μm is concentrated to low scattering angle. On the other hand, the scattered light intensity from a small diameter particle less than 1 μm is scattered wide angle. In order to capture the scattering light from the small particle size to the large particles size, modified Fourier optics shown in Fig. 1(a) is used. The Fourier optic is observed the low angle scattering pattern. A series of independent detectors, which are placed around the particles, are used for observing the high angle scattering pattern. A schematic of optics by the LS method, in which a Fourier transform lens is placed between the particles and an array detector, is shown in Fig. 1(a). The scattering light from the single spherical particle illuminated by a collimated laser beam gives rise to an angular variation of the intensity in the far field¹⁰, ¹⁶, ¹⁷ as shown in Fig. 1(b). A shape of the array detector has a concentric ring structure, because a shape of the particles is assumed to be spherical by Mie scattering model. Each detector has a finite dimension, covering some an angular range Δθ and Δφ, respectively. Values of Δθ and Δφ depend on a radial location of the detector. The total scattering intensity detects at average scattering angle θ and polar

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angle $\phi$ from particles in a scattering volume per unit detector area. All the concentric rings in this study have the same polar angles $\phi_1$ and $\phi_2$.

III. INVERSION PROBLEM

The relationship between PSD and angular dependence of the scattering intensity at each angle is expressed by the following the first kind Fredholm integral equation:

$$g(\theta) = \int_{D_{\text{max}}}^{D_{\text{min}}} K(n_r, \theta, D) f(D) dD,$$

where $f(D)$ is a frequency distribution of particle diameter $D$, $g(\theta)$ is scattering intensity by $f(D)$ at an angle $\theta$, and $K(n_r, \theta, D)$ is relative scattering intensity at the angle $\theta$ with the particle size $D$ for a given relative refractive index of particle $n_r$. The PSD must be solved by the inversion method with the theoretical scattering model $K(n_r, \theta, D)$ and the intensity of the scattering pattern $g(\theta)$.

If values of the PSD, the intensity of scattering light at the angle $\theta$, and the relative refractive index $n_r$ are approximated by discretization, the integral equation of Eq. (1) is transformed into a set of linear algebraic expression as follows:

$$g_i = \sum_{j=1}^{m} K_i(D_j) f(D_j) \Delta D_j,$$

where $g_i$ is a scattering intensity belonging to $i$th detector, $f(D_j)$ is a frequency size distribution of diameter $D_j$ that belongs to $j$th class, and $m$ is the maximum number of columns of particle size classes. Kernel function $K_i(D_j)$ is calculated from Mie scattering model with the particle size $D_j$ at $i$th detector for the given relative refractive index of particle $n_r$. If the total number of detectors is $n$, kernel function $K_i(D_j)$ is an $m \times n$ matrix. The physical meaning of $K_i(D_j)$ is relative scattering intensity at $i$th detector with the diameter of the particle $D_j$.

When the size distribution $f(D_j)$ is known, the scattering intensity $g_i$ can be calculated from Eq. (2). In order to obtain an unknown PSD, an inversion problem from Eq. (2) must be solved. Twomey reported a robust, nonlinear iteration inversion method to solve PSD. The algorithm uses the kernel function $K_i(D_j)$ to stabilize the solution of PSD. Moreover, the matrix of the kernel function is not required to be a square matrix by the Twomey nonlinear iteration method. Therefore, the number of detectors does not determine the number of columns of particle size classes, and the kernel function is used to calculate the PSD. The result of the Twomey iteration method is sensitive to a response of the kernel function. In order to obtain a stable calculation result of PSD, the MNLI method is used exclusively to estimate the PSD in this study.

The MNLI process is shown in Fig. 2

![Diagram](image)

FIG. 2. A schematic flow of the MNLI method.

- Initial guess $f^0(D_j)$
- Calculating $(r_i, p^{-1}) K_i(D_j)$ based on Eq.(3)
- Next iteration
- Correct $f^{p-1}(D_j)$ to obtain $f^{p}(D_j)$ based on Eq. (4)
where \( p \) is the number of iterations, \( f^{(p)}(D_j) \) is calculated frequency size distribution of diameter \( D_j \) that belongs to \( j \)th class at \( p \)th iteration, \( S_i \) is the maximum value of \( i \)th series of \( K_i(D_j) \), and \( g_i \) is an observed scattering intensity belonging to \( i \)th detector. \( K'_i(D_j) \) is not zero values. \( K'_i(D_j) \) is less than unity. Eq. (4′) defines a correction factor \( r_i^{(p-1)} \). Initial particle size distribution \( f^{(0)}(D_j) \) is defined to start the calculation. The \( (r_i^{(p-1)}-1)K'_i(D_j) \) is calculated taking into account all particle sizes. Then, size classes are recalculated by Eq. (3) with the weighted function of \( (r_i^{(p-1)}-1)K'_i(D_j) \) for each detector, and the result is in a new PSD. Value of \( r_i^{(p-1)} \) makes large contributions to correct the PSD at the first a few iterations. As the number of iterations is increased, value of \( r_i^{(p-1)} \) tends to unity.

Each detector has a different response to the scattered light from the different particle sizes. An example of the response scattering function of 21 and 41 elements detector at 1.19 of \( n_r \) is shown in Figs. 3(a) and 3(b), respectively. The characteristic of the scattering response function is normalized to the maximum values of each \( i \)th detector in Fig. 3. The largest diameter of particle size is observed by a detector at the minimum scattering angle. The smallest diameter of particle size is observed by a detector at the maximum scattering angle. The detector at the maximum scattering angle observes the smallest diameter of particle. All scattering functions are calculated by Mie scattering model. Comparing Fig. 3(a) with Fig. 3(b), shape of each scattering function with 21 elements detector is close with 41 elements detector. Average scattering angle of 0.0007 rad. is 1st detector in Figs. 3(a) and 3(b), respectively. Average scattering angle at 5th of 21 elements detector and the 9th of 41 elements detector is 0.002 rad. When the number of detectors is increased, the number of overlapped scattering function curves at the same particle diameter is increased. However, the shape of the scattering function is not so much changed.

### TABLE I. Dimension and scattering angle of the 18 elements array detector.

<table>
<thead>
<tr>
<th>No.</th>
<th>Inner radius (( \mu )m)</th>
<th>Outer radius (( \mu )m)</th>
<th>Average scattering angle (rad.)</th>
<th>No.</th>
<th>Inner radius (( \mu )m)</th>
<th>Outer radius (( \mu )m)</th>
<th>Average scattering angle (rad.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>66</td>
<td>86</td>
<td>0.000711</td>
<td>10</td>
<td>1473</td>
<td>2052</td>
<td>0.0164</td>
</tr>
<tr>
<td>2</td>
<td>98</td>
<td>123</td>
<td>0.00104</td>
<td>11</td>
<td>2064</td>
<td>2928</td>
<td>0.0232</td>
</tr>
<tr>
<td>3</td>
<td>135</td>
<td>177</td>
<td>0.00146</td>
<td>12</td>
<td>2940</td>
<td>4110</td>
<td>0.0328</td>
</tr>
<tr>
<td>4</td>
<td>189</td>
<td>251</td>
<td>0.00206</td>
<td>13</td>
<td>4122</td>
<td>5862</td>
<td>0.0465</td>
</tr>
<tr>
<td>5</td>
<td>263</td>
<td>361</td>
<td>0.00291</td>
<td>14</td>
<td>5874</td>
<td>8226</td>
<td>0.0657</td>
</tr>
<tr>
<td>6</td>
<td>373</td>
<td>509</td>
<td>0.00412</td>
<td>15</td>
<td>8238</td>
<td>11730</td>
<td>0.0931</td>
</tr>
<tr>
<td>7</td>
<td>521</td>
<td>728</td>
<td>0.00582</td>
<td>16</td>
<td>11742</td>
<td>16459</td>
<td>0.132</td>
</tr>
<tr>
<td>8</td>
<td>740</td>
<td>1023</td>
<td>0.00822</td>
<td>17</td>
<td>16471</td>
<td>23466</td>
<td>0.188</td>
</tr>
<tr>
<td>9</td>
<td>1055</td>
<td>1461</td>
<td>0.0116</td>
<td>18</td>
<td>23478</td>
<td>35000</td>
<td>0.278</td>
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TABLE II. Catalog number and accuracy of the polystyrene latex samples.

<table>
<thead>
<tr>
<th>Catalog number</th>
<th>Nominal diameter (μm)</th>
<th>Accuracy (μm)</th>
<th>Catalog number</th>
<th>Nominal diameter (μm)</th>
<th>Accuracy (μm)</th>
</tr>
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<tr>
<td>3200A</td>
<td>0.199</td>
<td>0.006</td>
<td>4250A</td>
<td>49.7</td>
<td>0.7</td>
</tr>
<tr>
<td>3500A</td>
<td>0.499</td>
<td>0.005</td>
<td>4270A</td>
<td>68.6</td>
<td>0.8</td>
</tr>
<tr>
<td>4010A</td>
<td>1.02</td>
<td>0.022</td>
<td>4311A</td>
<td>113</td>
<td>1.6</td>
</tr>
<tr>
<td>4205A</td>
<td>4.99</td>
<td>0.04</td>
<td>4324A</td>
<td>239</td>
<td>4.8</td>
</tr>
<tr>
<td>4210A</td>
<td>10.00</td>
<td>0.05</td>
<td>4330A</td>
<td>300</td>
<td>6</td>
</tr>
<tr>
<td>4230A</td>
<td>30.1</td>
<td>0.22</td>
<td>4340A</td>
<td>398</td>
<td>8</td>
</tr>
</tbody>
</table>

IV. COMPUTER SIMULATION

A computer simulation used for observing the influence of the number of detectors is broken into two steps. First, simulated scattering intensity distribution (input data) is generated according to Eq. (2) for a given PSD and the number of detectors. Second, the input data are processed by the MNLI method, and then the calculated PSD is compared with the given PSD.

The computer simulation can show how well input data are converted under the ideal conditions. The input data are generated by Mie scattering model at a wavelength of 633 nm and a relative refractive index of 1.19 or 1.33. The given PSD is calculated based on a log-normal distribution at five different mean diameters (MDs) in a range from 0.546 to 214 μm at standard deviation (SD) at 0.27 and 0.68, respectively. The number of detectors is changed in a range from 6 to 81. The detectors are capable of collecting the scattered light over a range of four orders of scattering angle θ from 0.0007 to 2.5 rad. 0.785 rad. for φ1, and 2.36 rad. for φ2, which correspond to the concentric structure of the array detector, are utilized for the array detector. The effect of a noise on the scattering intensity is not carried out, because the noise did not hardly influenced the MNLI method in the author's previous study.14

To examine a convergence criteria for the calculated PSD, a residual square of difference between the given PSD and the calculated PSD at each column of particle size $r^2$ is used

$$r^2 = \sum_{j=1}^{m} (f_c(D_j) - f_o(D_j))^2,$$

where $f_c(D_j)$ is the calculated PSD, and $f_o(D_j)$ is the given PSD. When the calculated PSD is closed to the given PSD, value of $r^2$ is reduced.

V. EXPERIMENTAL METHOD

The optical layout of the experimental setup is sketched in Fig. 1(a). A wavelength of the light source for the experiment is 633 nm. Three kinds of optical systems have 21, 41, and 81 elements detector, respectively. Each Fourier optic consists of 18 and a 36 or 75 elements detector, respectively. A focus length of the lens is 106 mm for the Fourier optics. The array detectors have the same concentric configuration. An overview of 18 elements photodiode array detector, for example, is shown in Fig. 4(a). A close-up of a center of the photo diode array is shown in Fig. 4(b). The radius of the array

![Fig. 5](http://rsi.aip.org/about/rights_and_permissions)

FIG. 5. Comparison between a given PSD (solid line) and a calculated PSD (●) at MD of 0.546 μm, 2.77 μm, 55.1 μm, and 214 μm with (a) 6 elements detector, (b) 21 elements detector, and (c) 81 elements detector, respectively. MD of all PSD characterized by the same log-normal size distribution with SD of 0.27. The test is performed with the different number of detectors. Q3(Dx) is cumulative size distribution based on mass, and longitudinal axis has a logarithmic scale.
FIG. 6. Comparison between a given PSD (solid line) and a calculated PSD (●) at 0.68 of SD. The other conditions are same as Fig. 5.

detector Rd, which has a concentric structure, is 35 mm maximum. The detector is divided into 18 elements logarithmically surrounding the optical axis. The scattered light is averaged over the solid angle associated with individual elements detector. Table I shows dimensions and average scattering angles of the 18 elements array detector. Scattering angles of the independent detector in the case of the 21 detectors optics are 0.524 rad., 1.23 rad., and 2.41 rad., respectively.

Twelve kinds of traceable mono-dispersed polystyrene latex (PSL) samples, whose size range is from 0.199 to 398 μm, are used for evaluating the accuracy of calculated results with various detectors. PSL samples are manufactured by Thermo Fisher Scientific Ltd. The accuracies of the PSL are shown in Table II. A non-spherical alumina sample (Catalog number: WA-240, Fujimi Co. Ltd.) is used for observing a result of real sample. A relative refractive index of 1.19 is used for the PLS samples, and 1.33 is used for the alumina sample to calculate PSD, respectively. The experimental samples are suspended in water and circulated in the cell not to be segregated. The samples are measured at around 80 transmitted percent.

VI. RESULTS AND DISCUSSION

Typical results of the computer simulation are illustrated in Fig. 5 with SD of 0.27 at various MD. $Q_3(D_x)$ is cumulative size distribution based on mass as follows:

$$Q_3(D_x) = \sum_{i=1}^{N} f(D_j) \Delta D_j.$$  

(6)

The number of detectors is varied from 6 to 81. A solid line indicates given PSD, and symbol of filled circle indicates calculated PSD. When the number of detectors is 6, the cal-

FIG. 7. Relation between $r^2$ and the number of detectors by computer simulation. The given PSD is the log-normal distributions at (a) SD of 0.27 and (b) SD of 0.68, respectively.
The number of physical detectors [-]

FIG. 8. Relation between RN and the number of physical detectors and the number of physical detectors. The result at MD of 0.546 μm is shown by symbol (●), at MD of 2.72 μm is shown by symbol (▲), at MD of 55.1 μm is shown by symbol (▼), and at MD of 214 μm is shown by symbol (■), respectively. Value of SD is 0.27, and discrimination level is 3%.

culated PSD is not agreed with the given PSD as shown in Fig. 5(a). When the number of detectors is increased, the given PSD and the calculated PSD show reasonable agreement.

Fig. 6 shows results of the computer simulation at SD of 0.68. The other conditions are the same as Fig. 5. The calculated results show reasonable agreement with 6, 21, and 81 elements detector, respectively. The calculated PSD with 6 elements detector is not agreed with the given PSD at MD of 214 μm. The results of the computer simulation show that a wide PSD (SD = 0.68) from 6 to 81 elements detector is in better agreement than a narrow PSD (SD = 0.27).

The influence of the number of detectors is evaluated by the residual square $r^2$ which is defined by Eq. (5) by the computer simulation. The number of detectors is varied in a range from 6 to 81, and MD is varied in a range from 0.546 to 214 μm, respectively. The results of computer simulation with the narrow PSD (SD = 0.27) are shown in Fig. 7(a), and simulation results with the wide PSD (SD = 0.68) is shown in Fig. 7(b), respectively. The wide PSD (SD = 0.68) in Fig. 7(a) tends to less influence of the number of detectors than the narrow PSD (SD = 0.27) shown in Fig. 7(b). Values of $r^2$ with the narrow PSD decrease quickly as the number of detectors is increased. When the number of detectors is larger than 20, values of $r^2$ with the narrow PSD have a tendency to become plateau. Values of $r^2$ with the wide PSD show almost less dependency of the number of detectors than the narrow PSD.

The wide PSD has smoother scattering pattern than the narrow PSD. It is easy to expect that the smooth scattering pattern requires less number of detectors than sharp scattering pattern to solve the inverse problem. From these results, the wide PSD is less dependency of the number of detectors than the narrow PSD for the LS method. When PSD is narrow, values of $r^2$ at MD of 0.546 μm show less dependency of the number of detectors than at MD of 2.72 μm and more.

As particle size is large, a scattering signal is focused on detectors for small angle as shown in Fig. 2. On the other hand, as particle size is small, a scattering signal is observed on detectors for all angles. Authors expect that detectors, of which scattering signal is more than a discrete signal level, contributes only the calculation of PSD. As a scattering pattern is focused on a limited number of detectors, the number of useful detectors is smaller than the number of physical detectors. To observe the influence of the number of useful detectors, a ratio of the number of useful detectors and the number of physical detectors RN is defined by the following equation:
equation:
\[ RN = \frac{I_e}{I_p}, \tag{7} \]
where \( I_e \) is the number of useful detectors, and \( I_p \) is the number of physical detectors. 0.3% of discrete signal level, which is obtained from the experimental setup, is adopted. As the number of physical detectors is increased, the number of useful detectors is also increased. Not all detector elements contain useful information about the calculation of PSD. Fig. 8 shows the relation between RN and the number of physical detectors with SD of 0.27 at MD of (a) 0.18 \( \mu \)m, (b) 2.72 \( \mu \)m, (c) 55 \( \mu \)m, and (d) 280 \( \mu \)m, respectively. When the number of physical detectors is increased, value of RN is decreased with MD of 0.18 \( \mu \)m and 2.72 \( \mu \)m, respectively. Values of RN at MD of 0.546 \( \mu \)m is always less than the other conditions of RN. Values of RN at MD of 2.72 \( \mu \)m and MD at 280 \( \mu \)m are almost constant as the number of detectors is increased. The number of useful detectors is not so much increased, as the number of physical detectors at MD of 0.546 \( \mu \)m is increased. This result is in agreement with a tendency of \( r^2 \) values with the narrow PSD at MD of 0.546 \( \mu \)m. The relation between the number of useful detectors and the number of physical detectors can be explained by the relation between \( r^2 \) and the number of detectors when PSD is narrow. In this result of the computer simulation, the number of detectors over 20 is the minimum number of detectors by computer simulation.

Results of the experiment and computer simulation listed by the number of detectors are shown in Fig. 9. The relative error CV is defined by the following equation:
\[ CV = \frac{D_c - D_m}{D_c} \times 100, \tag{8} \]
where \( D_c \) is the certified sample or given mean size diameter, and \( D_m \) is the measured or calculated mean size diameter, respectively. The accuracies of the experimental results and the computer simulation fit within \(-4 \) to \(+6\)%. Influence of the number of detectors does not observe in a range of 21 to 81 elements detector. As the computer simulation results predicted, the effect of the number of detectors is quite weak with the experimental results from the number of 21 detectors.

Results of poly-disperse alumina sample with the different number of detectors are shown in Fig. 10. The experimental results at the condition of three difference number of detectors are within the usual limit of calculation. The calculated PSD for the alumina sample also shows less dependency on the number of detectors in a range of 21 to 81.

VII. CONCLUSION

The effect of the number of detectors on the inversion problem of the LS method has been studied by the computer simulation and experimentally method. The wide PSD (SD = 0.68) observed less effect of the number of detectors than the narrow PSD (SD = 0.27) except the result of narrow PSD at MD of 0.546 \( \mu \)m. When the detector elements are over 20, the influence of the number of detectors is decreased. The PSD at MD of 0.546 \( \mu \)m shows less dependency of the number of detectors than the other MD in the case of the narrow PSD. Useful detectors, which have a signal over the discrete level, make influence the calculated PSD. When MD is 0.546 \( \mu \)m with the narrow PSD, the influence of the number of detectors can be explained by the ratio of the number of useful detectors and the number of physical detectors in the case of the narrow PSD.

From the experimental results of mono-disperse polystyrene latex samples and alumina sample, influence of the number of detectors is not observed to compare the number of 21, 41, and 81 elements detector. Contribution to the calculated PSD is reduced when the number of detectors is 21 or more.

Preliminary results show the calculated PSD will be unsusceptible from 21 elements detector by the computer simulation and the experimental method.

![Graph](https://via.placeholder.com/150)

**FIG. 10.** Results of measured aluminum sample with three different number of detectors.