METHODS UTILIZING THE PHENOMENA OF LIGHT SCATTERING TO
MEASURE MICROSCOPIC ALUMINUM OXIDE PARTICLE SIZE IN BOTH
SOLUTION AND TURBULENT AIR FLOW

by
Lawrence Edward Deese II

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Approved by

Advisor: Dr. Nathan Murray

Reader: Dr. Adam Green

Reader: Dr. Adam Smith
ABSTRACT

Lawrence Deese: Methods Utilizing the Phenomena of Light Scattering to Measure Microscopic Aluminum Oxide Particle Size in Both Solution and Turbulent Air Flow

The purpose of this research project was to measure microscopic aluminum oxide (alumina) particle size both in solution and in air flow. Experimentally, the process of sizing microscopic particles is technically challenging. Two methods were used for measuring the sizes of 4 different manufacturer labeled aluminum oxide particles sizes. The first method was using Dynamic Light Scattering analyzed by a LS Spectrometer to measure the alumina particle size while suspended in solution. The second method was using the forward angle scattering transmissometer (FAST) technique to measure alumina particle size while subjected to a turbulent air flow. Accurately measuring these particle sizes, with the aim of confirming the manufacturer labeled size, ultimately helps validate experiments which use these particles performed at the National Center Physical Acoustics (NCPA).
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1.0 INTRODUCTION TO DYNAMIC LIGHT SCATTERING (DLS)

1.1 Overview of DLS

While suspended in solution, macromolecules are in constant interaction with solvent molecules. These interactions result in random motion of the macromolecule, more commonly known as Brownian motion. Malvern Instruments provides a brief introduction to dynamic light scattering, a technique that uses the natural Brownian motion to determine particle size. To determine particle size by Brownian Motion analysis it is necessary to determine the translational diffusion coefficient (D), which can be characterized as the speed of diffusion or Brownian Motion. The larger the particle, the slower the Brownian motion will be and vice versa. Certain properties of solutions must be known for Dynamic Light Scattering to produce valid results. For example, the temperature of the solution must be accurately known in order to characterize solution viscosity. Temperature must also be held stable or else convection currents in the sample will cause non-random movements that will alter the measurements for the translational diffusion coefficient, ultimately corrupting the correct interpretation of particle size (Malvern Instruments Ltd).

1.2 Hydrodynamic Diameter

The hydrodynamic diameter calculated from the Stokes-Einstein equation is the diameter of a hard sphere with the same translational diffusion coefficient as the
particle of interest. Therefore, the measurement assumes a smooth surface structure and spherical particle geometry.

A particle’s hydrodynamic diameter can be calculated from the translational diffusion coefficient utilizing the Stokes Einstein equation

\[ d(H) = \frac{kT}{3\pi \eta D} \]  

(1)

where

- \( d(H) \) = Hydrodynamic diameter,
- \( D \) = Translational diffusion coefficient,
- \( k \) = Boltzmann’s constant,
- \( T \) = Absolute temperature, and
- \( \eta \) = Viscosity of the solution.

\( T \), \( k \), and \( \eta \) should all be known constants for the solution being tested. Thus, once the translational diffusion coefficient is experimentally determined, the hydrodynamic diameter can be calculated.

1.3 DLS Principle

For DLS to perform particle sizing analysis properly, it must measure the speed at which the particles diffuse in solution due to Brownian motion. This speed is known as the translational diffusion coefficient and DLS determines this coefficient by measuring
the rate at which the intensity of scattered light fluctuates when detected using an appropriate optical setup (Malvern Instruments Ltd).

Fluctuating light intensity data is obtained and analyzed by auto-correlation, which processes the spectrum of intensities and measures the degree of similarity a signal has with itself at very small time scales. According to Malvern Instruments, if the particles are large, the signal will be change slower and correlation will persist longer than if the particles are small.

The Time Auto Correlation function is defined as

\[ G(\tau) = \frac{\langle I(t_0) \ast I(t_0 + \tau) \rangle}{I(t_\infty)^2} \]  

(2)

where

\[ G(\tau) = \text{Time Auto Correlation function}, \]

\[ I(t_0) = \text{Measured Laser intensity at time} = 0, \]

\[ \tau = \Delta t = \text{Time lag at very small time scales}, \]

\[ I(t_\infty) = \text{Measured Laser intensity at time} = \infty. \]

It should be well understood that since these measurements are taking place at very small time scales (nano-seconds), the \( I(t_\infty) \) term is the laser intensity averaged over a few seconds. Relative to the time scale however, this is an immense amount of time.

By plotting this correlation function and analyzing how this correlation decays over time:

\[(t + \Delta t, t + 2\Delta t, \ldots, t + \lim_{n \rightarrow \infty} n\Delta t),\]
information on suspended particle size can be determined (Malvern Instruments Ltd, p.3).

1.4 Correlation Function

To determine suspended particle size, the auto-correlation function is modeled with regard to the Stokes-Einstein equation. The correlation function behaves as an exponential decaying curve, it can be modeled by the equation

\[ G(\tau) = A(1 + B e^{-2\Gamma \tau}) \]  

(3)

where

\[ A = \text{Baseline of the Time Auto Correlation Function}, \]
\[ B = \text{Intercept of the Time Auto Correlation Function, and} \]
\[ \Gamma = D q^2. \]

The decay coefficient, \( \Gamma \), includes the scattering vector defined by the term \( q \), which depends on the configuration of the measurement and the particle’s refractive index according to

\[ q = \left( \frac{4 \pi n}{\lambda_0} \right) \sin \left( \frac{\theta}{2} \right) \]  

(4)

where

\[ n = \text{Refractive index of particle}, \]
\[ \lambda_0 = \text{Wavelength of the laser light, and} \]
\[ \theta = \text{Scattering angle of measured laser light}. \]
Because the above parameters are held constant throughout the measurement process, $q$ is also constant. Therefore, the Translation Diffusion Coefficient, $D$, can be directly calculated from $\Gamma$. Finally, $D$ can be used with the Stokes-Einstein equation (eq. 1), and a mean hydrodynamic diameter of particles suspended in solution can be calculated.

Occasionally, solution samples of particles will be polydisperse. In this case, the time auto correlation function obtained from a DLS analysis will be the sum of numerous exponential decay functions that decay at different rates corresponding to each particle size in the solution. By fitting a multiple exponential to the polydisperse auto correlation function, the range of particle sizes in solution becomes known. Then, by analyzing the relative intensity of light scattered from different particles in this size range allows for a particle size distribution of the polydisperse solution to be obtained (Malvern Instruments Ltd, p.5).
2.0 EXPERIMENTS PERFORMED USING LS SPECTROMETER

2.1 Introduction to LS Spectrometer

The LS Spectrometer is a particle sizer made by LS Instruments which is capable of performing both Static Light Scattering (SLS) and Dynamic Light Scattering (DLS). The instrument automates the measurement process described in Section 1.0. The setup also enables characterization in the form of hydrodynamic radius, the radius of gyration, form and structure factors, size distribution, as well as molecular weight for particle sizes ranging from 0.15 nanometers to 0.5 micrometers. The system schematics are shown in Figures 1 and 2 for reference.

Figure 1. LS Spectrometer. (LS Spectrometer, n.d.).
2.2 Steps for successful solution preparation

It should be noted that much time and many trials took place before obtaining the correct particle size data using the LS spectrometer. Particle agglomeration poses a significant issue when using light scattering for the purposes of particle sizing. Therefore, solution preparation is vitally important in assuring correct data can be obtained.

The preparation of initial alumina test solutions followed a proportion of 2.3g alumina / 266.25mL ethanol. To ensure the particles were sufficiently dispersed
while suspended in solution, all solutions were sonicated (for breaking up small clumps of particles) and treated with a small amount of HCl (limit the effect of electrostatic force interactions between particles). However, despite these treatments, data obtained from the LS spectrometer still showed particles to be bigger than what they were manufacturer labeled.

Assuming agglomeration to still be the leading cause, sonication and HCl treatments were adjusted in duration and quantity respectively. In addition to these adjustments the surfactants CTAB and SDS were included in certain tested solutions to reduce solution viscosity. However, due to their chemical nature, once these surfactants were introduced into alumina solutions, micelles formed around clumps of the suspended alumina. This process essentially facilitated agglomeration and hindered efforts of successful particle dispersion.

The effect of multiple scattering was inevitably determined to be the reason for incorrect data obtained by the LS spectrometer. The effect of multiple scattering corresponds directly with solution opaqueness. Therefore, to limit the effects of multiple scattering a much smaller mass of alumina was used in solution preparation.

Ultimately concentrated alumina solutions treated with acid and sonication likely did not effect LS spectrometer recorded particle size due to particle agglomeration, but did effect LS Spectrometer recorded particle size due to multiple scattering. In order to create a solution transparent enough to limit the effects of multiple scattering, a very small amount of alumina was used in tested solutions. The final successful solution is listed in Table 1, and it was held constant for each particle size reported in section 2.5.
Table 1. Steps for successful solution preparation.

<table>
<thead>
<tr>
<th>PARTICLE SIZE</th>
<th>STEP 1: AMOUNT OF ALUMINA</th>
<th>STEP 2: AMOUNT OF 190 PROOF ETHANOL</th>
<th>STEP 3: AMOUNT OF 0.1M HCL</th>
<th>STEP 4: TIME OF SONICATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05 micron</td>
<td>&lt; 0.01g</td>
<td>50mL</td>
<td>1mL</td>
<td>7 minutes</td>
</tr>
<tr>
<td>0.3 micron</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 micron</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3 micron</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

2.3 Steps for using the LS Spectrometer

Alumina particles of each previously mentioned size were measured while suspended in ethanol solution using a single angle measurement with the LS Spectrometer. The steps for using the LS Spectrometer for alumina particle sizing are as follows:

1. Turn power on to main unit and water bath, and open LS Spectrometer program.
2. While in the program click the ‘change sample’ button
3. Unscrew top to sample chamber and remove sample if one is already present
4. Syringe approximately 2mL of sample into a Kimble borosilicate Disposable Culture tube 10mm(diameter) X 75mm (length) and cap the top with wax peri-film.
5. Wash the outside of the cubet with pure ethanol and then carefully place the cubet into the sample chamber
6. Screw the top back onto the sample chamber
7. Click ‘ok’ on the computer program to indicate that changing the sample has successfully been completed
8. Change the scattering geometry to 2D

9. Click on ‘set up script’ and change the following
   a. Start angle to 90° and final angle to 90°,
   b. Step size to 0.0
   c. Number of measurements to 1
   d. Run time to 100 seconds

10. After completing the script click ‘Insert Line’, and create a folder for the data to save to once the measurement has completed

11. Click ‘measurement’ tab, then click ‘Start Script’
2.4 Data attained from LS Spectrometer

The data attained from the LS spectrometer is plotted in Figure 3, which shows the Probability Density Function, (PDF), for each particle size.

![Figure 3](image)

**Figure 3.** Particle size distribution data obtained from LS Spectrometer ($\mu =$ peak of main maxima for each distribution).

Considering the peak of each PDF, the results show that each of the four alumina particle sizes tested in solution are quite comparable to their respected manufacturer labeled size. The difference between the claimed size and measured size increases with the nominal size. This is likely due to the highly non-spherical nature of the particles, which is also likely responsible for the minor peaks in the PDF of the 1$\mu$m and 3$\mu$m particles. Therefore, based off these results, the assumption of the alumina particles being spherical becomes more problematic as the claimed particle size increases.
3.0 INTRODUCTION TO THE FORWARD-ANGLE-SCATTERING-TRANSMISSOMETER (FAST) TECHNIQUE

3.1 Overview of FAST technique

The FAST technique is a method for determining particle parameters while suspended in media using light scattering data. The DLS method is not suitable for obtaining particle size measurements for particles diffusing in a high-speed gas flow. Therefore, an effort was made to adapt the Forward-Angle-Scattering-Transmissometer technique previously described by Nefedov, (1998).

The FAST technique has been successful in determining mean particle sizes of monodisperse and polydisperse solutions (Nefedov, 1998). The experimenters in the Nefedov paper were able to measure the size of polystyrene spheres distilled in water using the FAST technique. The aim of this experiment is to use the FAST technique to measure alumina particle size distribution while the particles are subjected to turbulent air flow in a high-speed stream.
3.2 Experimental setup

A basic optical setup for the FAST technique is shown below in Figure 4.

**Figure 4.** Basic optical scheme for FAST setup based off Nefedov experiment.

List of parts for figure 4:

- 1 = laser source
- 2 = sample space with particles of radius $r$
- 3 = aperture diaphragm of varying diameters: 5.0, 7.6, 11.7, 17.3, 26.0, 39.5, 59.5 and 75.0mm on a rotating disk
- 4 = lens condenser
- 5 = reference photodiode, $PD_r$ (reference)
- 6 = post scatter photodiode, $PD_a$ (auxiliary)
- 7 = post aperture photodiode, $PD_b$ (basic)
- $l$ = width of particle sample space
- $L$ = distance between particle sample space and aperture diaphragm
3.3 Brief discussion on Light Scattering

Light scattering occurs once incident light becomes redirected after interacting with small particles or molecules in a medium. There are different forms of light scattering that can occur depending on the factors of light wavelength and particle radius. Using the equation

\[ x = \frac{2\pi r}{\lambda} \]  

(5)

where

\[ r = \text{radius of the particle, and} \]
\[ \lambda = \text{wavelength of light,} \]

the type of light scattering can be predicted based on the calculated value of \( x \).

\( x \) is a non-dimensional size parameter where if

\[ x << 1 \quad \text{Rayleigh scattering occurs,} \]
\[ x \sim 1 \quad \text{Mie scattering occurs, and} \]
\[ x >> 1 \quad \text{Geometric scattering occurs.} \]

A chart that gives the ranges of the types of light scattering depending on these factors is displayed in Figure 5 shown below.
Figure 5. Scattering regimes depending on particle size and wavelength.

(Scattering Lecture, n.d.).

Given that the laser light source used in this experiment has a 633nm wave length, the 0.3, 1, and 3 micron alumina particle sizes certainly fall within the range in which Mie scattering occurs. However, due to its size and the wavelength of laser light, the 0.05 micron alumina particle size looks to be on the threshold of the Mie scattering regime. Because of this, the 0.05 micron alumina particle will likely present characteristics of both Mie and Rayleigh light scatter.
3.4 Theoretical Background

The approach of (Nefedov, 1998) considers a transparent non-absorbing medium with width ($l$) and inserted inside is a suspension of non-absorbing homogenous spherical particles with a radius ($r$) and number density ($N_p$). According to Nefedov, if these particles are struck by a collimated laser beam, the Transmittance ($T$) of the particle suspension can be described as

$$T \equiv \frac{l}{l_0} = e^{-\tau}$$

where

$$\tau = N_p \bar{\sigma}_{ext} l,$$

$$l_0 = \text{incident light intensity},$$

$$l = \text{transmitted light intensity}, \text{ and}$$

$$\bar{\sigma}_{ext} = \text{mean extinction cross section of the suspended particles}.$$

The mean extinction cross section of the particles $\bar{\sigma}_{ext}$ can be defined as

$$\bar{\sigma}_{ext} = \int_0^\infty \sigma_{ext} f(r) dr$$

(7)

where $\sigma_{ext} = \sigma_s + \sigma_a$. $\sigma_{ext}$ is the extinction cross section of a single particle of radius $r$. $\sigma_s$ and $\sigma_a$ are the scattering and absorption cross sections for a single particle, and $f(r)$ is the particle size distribution function (Nefedov, 1998).
Now define the geometric path length for the sample volume of particles to be \( l \), and the distance between the sample volume of particles and the aperture diaphragm to be \( L \). According to Nefedov, by adhering to the ratio \( \left( \frac{l}{L} \right) \leq 0.1 \) in the experimental setup, the sample volume of particles will be short enough to properly measure scattered light intensity.

According to Nefedov, if we consider that some of this scattered light at angles \( \theta \leq \theta_d \) enters PD\( b \), where \( \theta_d \) = polar angle of the solid angle subtended by the PD\( b \), then the theoretical integral cross section \( \sigma^*(\theta_d) \) measured by the photodetector can be defined as

\[
\sigma^*(\theta_d) = \int_0^\infty \left[ \sigma_{\text{ext}} - \left( \frac{1}{2} \right) \sigma_{\text{sca}} \int_0^{\theta_d} p_\theta \sin \theta d\theta \right] f(r)dr
\]

where \( p_\theta \) = the single-particle scattering phase function, and \( \theta \) = scattering angle of light. Considering the scenario when the number density \( (N_p) \) of particles is unknown, the scattered light intensity measurement recorded will yield a relative value of the integral cross section measured by the photodetector \( \sigma^*(\theta_d) \) (Nefedov, 1998). By measuring \( \sigma^*(\theta_d) \) at different detector angles, \( \theta_d \leq 10^\circ \), certain optical properties of monodisperse solutions can become known (Nefedov, 1998). Varying the angle for which scattered light is measured by PD\( b \) can be achieved by collecting the scattered light through eight different aperture sizes. According to Nefedov, by measuring light intensities at distinct points in the experimental setup, an experimental angular distribution function, \( q^{\text{meas}}(\theta_{dl}) \), can be obtained.
\[ q^{\text{meas}}(\theta_d) = \frac{\ln \left( \frac{I_b^0}{I_r^a} \right) - \ln \left( \frac{I_b^0}{I_r^b} \right)}{\ln \left( \frac{I_a^0}{I_r^a} \right) - \ln \left( \frac{I_a^0}{I_r^b} \right)} \]

where the subscript \( i \) denotes one of the 8 different apertures. The superscript 0 on \( I \) denotes the laser intensity recorded when no particles were present in the medium. The subscripts \( a, b, \) and \( r \) on \( I \) denote the laser light intensities recorded at their respective photodiodes shown in section 3.2.

The angular distribution function, \( q(\theta_d) \), is significant since the extinction cross section, \( \sigma_{\text{ext}} \), and integral cross section, \( \sigma^*(\theta_d) \), are related to it by the formula

\[ q(\theta_d) \equiv \frac{\sigma^*(\theta_d)}{\sigma_{\text{ext}}} \]

A theoretical angular distribution function, \( q^{\text{calc}}(\theta_d) \), can be obtained by the equation:

\[ q^{\text{calc}}(\theta_d) = \frac{\sigma^*(\theta_d)^{\text{calc}}}{\sigma_{\text{ext}}^{\text{calc}}} = \left( \int_{0}^{\infty} \left[ \sigma_{\text{ext}} - \frac{1}{2} \sigma_{\text{sca}} f^d \right] \sigma_{\text{ext}}^{\text{calc}} f(\mathcal{r}) d\mathcal{r} \right) \]

where the theoretical extinction cross section, \( \sigma_{\text{ext}}^{\text{calc}} \), and single particle scattering phase function, \( p_\theta \), are calculated using Mie theory. To solve \( q^{\text{calc}}(\theta_d) \) completely, a formula to describe the particle size distribution function, \( f(\mathcal{r}) \), is also required. This function can be modeled by many different formulas, however, a mathematical model with Gaussian form would be most appropriate. For the purpose of this experiment, a mathematical model will be generated from the particle distribution function attained by
the LS spectrometer for each particle size. This mathematical model should take the form of

\[ f(r) = ae^{-(\frac{r-b}{c})^2} \]  

(12)

where \( a, b, \) and \( c \) are free parameters. Equation 12 can then be fitted with the DLS data by varying the previously mentioned parameters using a best fit approach.

Therefore, once experimental measurements are made, the measured angular distribution functions can be generated and plotted for each particle size. Once alumina’s theoretical extinction cross section and single-particle-scattering phase function are calculated from Mie theory, a theoretical angular distribution function can be calculated and plotted. These two functions will be compared to one another and if they are the same, then manufacturer labeled alumina particle size will be validated.

### 3.5 Introduction to Mie Theory

Christian Mätzler explains the mathematics involved in Mie theory in his research report “MATLAB Functions for Mie Scattering and Absorption”. According to Mätzler, in order to solve for light scattering particle parameters using Mie theory, the assumption of a homogenous sphere must be made for the particle. Then the Mie coefficients \( a_n, b_n, c_n, d_n \), which are important values that Mie calculations heavily rely on, can be solved for. The Mie coefficients \( a_n, b_n \) describe the amplitudes of light in a field external of the particle, while \( c_n, d_n \) describe these amplitudes for the particle’s internal field. The radiative transfer aspect of Mie Scattering, which occur in the external particle field,
depend on the Mie coefficients $a_n, b_n$, but not on $c_n, d_n$. The Mie coefficients $c_n, d_n$ are needed when the particle internal electric field is of interest, for example if the particle significantly absorbs light radiation.

All substances have an optical constant named the complex index of refraction

$$m = n - i\kappa$$

(13)

where

- $n = \text{index of refraction (real part)}$, and
- $\kappa = \text{index of absorption (imaginary part)}$.

Light extinction by a particle is determined by both these elements. Therefore, if a particle significantly absorbs light radiation, it becomes necessary to know the imaginary part of the refractive index in order to obtain accurate Mie calculated parameters.

Nefedov states in (Nefedov, 1997, 1365) that alumina is a weakly absorbing particle. This assertion is supported by detailed studies conducted to determine the spectral absorption coefficient for alumina in the near infrared spectrum of light (Aleksey, 2016, 164). The spectral absorption coefficient, $\alpha$, is defined as

$$\alpha = \frac{4\pi\kappa}{\lambda}$$

(14)

where

- $\kappa = \text{index of absorption (imaginary part)}$, and
- $\lambda = \text{wave length of light.}$
A plot for alumina’s spectral absorption coefficient depending on temperature and light wave length is shown in Figure 6 below.

Figure 6. Spectral optical properties of alumina. (Aleksey, 2016).

The laser light used in this experiment is 633nm (0.63μm), and experiments will be performed at room temperature. Therefore, from the plot above, alumina’s spectral absorption coefficient will be 0.004 cm⁻¹ - 0.005 cm⁻¹. Using equation 13 to calculate the index of absorption utilizing the spectral absorption coefficient, one will find that alumina’s index of absorption is small enough to be considered insignificant in Mie scattering calculations. Therefore, the imaginary part of alumina’s complex refractive index is assumed 0 making the absorption of light energy by alumina negligible in these
experiments. Consequently, $a_n, b_n$ will be the only Mie coefficients of interest. These coefficients can be described mathematically by

$$a_n = \frac{m^2 j_n(mx)[xj_n(x)]' - \mu_1 j_n(x)[mxj_n(mx)]'}{m^2 j_n(mx)[xh_n^{(1)}(x)]' - \mu_1 h_n^{(1)}(x)[mxj_n(mx)]'}$$

$$b_n = \frac{\mu_1 j_n(mx)[xj_n(x)]' - j_n(x)[mxj_n(mx)]'}{\mu_1 j_n(mx)[xh_n^{(1)}(x)]' - h_n^{(1)}(x)[mxj_n(mx)]'}$$

(15)

where

$m = \text{refractive index of particle while suspended in medium},$

$x = \text{the size parameter defined by the equation } x = ka,$

$a = \text{particle radius},$

$k = \text{wave number defined by equation } k = \frac{2\pi}{\lambda},$

$\lambda = \text{wavelength of light in medium},$

$\mu_1 = \text{ratio of magnetic permeability of the particle to the magnetic permeability of the medium},$

$j_n(x) \text{ and } h_n^{(1)}(x) = \text{spherical Bessel functions of order } n \text{ (where } n = 1, 2, \ldots \text{) and }'$

$' = \text{derivatives of the functions in which they are operated on}$

where

$$[xj_n(x)]' = xj_{n-1}(x) - nj_n(x) \quad \text{and} \quad [xh_n^{(1)}(x)]' = xh_{n-1}^{(1)}(x) - nh_n^{(1)}(x).$$

The efficiencies $Q_i$ for which light interacts with a scattering particle of radius $a$ are the cross sections $\sigma_i$ normalized to the particle cross section, $\pi a^2$. 


\[
Q_i = \frac{\sigma_i}{\pi a^2}
\]  

(16)

where

\[i = \text{extinction (ext), absorption (abs), or scattering (sca)}.\]

Due to energy conservation,

\[
Q_{ext} = Q_{sca} + Q_{abs} \quad \text{and} \quad \sigma_{ext} = \sigma_{sca} + \sigma_{abs}.
\]

(17)

Since the absorption of light radiation by alumina is negligible in this experiment, \(Q_{abs}\) and \(\sigma_{abs}\) are both 0. This simplifies equation 12 and equation 13 to

\[
Q_{ext} = Q_{sca} \quad \text{and} \quad \sigma_{ext} = \sigma_{sca}
\]

(18)

\[
Q_{ext/sca} = \frac{\sigma_{ext/sca}}{\pi a^2}
\]

(19)

where

\[\sigma_{sca} = \text{scattering cross section of particle, and} \]

\[\pi a^2 = \text{particle cross section}.\]

\(Q_{sca}\) can be calculated using the Mie coefficients, \(a_n, b_n\), previously mentioned.

\[
Q_{sca} = \frac{2}{\lambda^2} \sum_{n=1}^{\infty} (2n + 1)(|a_n|^2 + |b_n|^2).
\]

(20)
Once the scattering efficiency is calculated, the extinction cross section can be solved for using

$$ Q_{\text{sca}} = \frac{\sigma_{\text{ext}}}{\pi a^2} $$

(21)

Another parameter used for deriving the theoretical integral cross section is the single-particle-scattering phase function, $p_\theta$. The phase function essentially describes the angular dependence of particle scattered light intensity (Frisvad, 2018). Models for the scattering phase function are shown in Figure 7 below.

**Figure 7.** Models of the scattering phase function at different size parameters given a certain light wavelength. (Scattering Lecture, n.d.)
This scattering phase function is important because when incident light at a specific wavelength strikes a particle with a certain size, the light will scatter at a precise angle corresponding with the particle’s particular size.

The scattering amplitude functions, $S_1$ and $S_2$, are critical to solving for the phase function because they describe the far field scattering of light and are defined as

$$S_1(\cos \theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \pi_n + b_n \tau_n)$$

(22)

and

$$S_2(\cos \theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \tau_n + b_n \pi_n)$$

(23)

where the functions $\pi_n(\cos \theta)$ and $\tau_n(\cos \theta)$ follow the recurrence relations,

$$\pi_n = \frac{2n-1}{n-1} \cos \theta \ast \pi_{n-1} - \frac{n}{n-1} \pi_{n-2} \quad \text{and} \quad \tau_n = n \cos \theta \ast \pi_n - (n+1) \pi_{n-1}$$

where

$$\pi_0 = 0; \quad \pi_1 = 1; \quad \pi_2 = 3 \cos \theta \quad \text{and} \quad \tau_0 = 0; \quad \tau_1 = \cos \theta; \quad \tau_2 = 3 \cos 2\theta.$$ 

If the incident light is unpolarized (Hovenier, 2004) and the assumption of a homogenous sphere is made for the particles, the formula for the phase function can be simplified (McLinden, 1999).
The simplified phase function is defined as

\[
p(\theta) = \frac{2\pi}{k^2 \sigma_{sca}} [ |S_1(\theta)|^2 + |S_2(\theta)|^2 ]
\]

(24)

where

\[ k = \text{wave number, and} \]
\[ \sigma_{sca} = \text{scattering cross section of particle.} \]

3.6 Obtaining scattering particle parameters from MatScat computer program.

A computer program by Jan Schäfer named “MatScat” has been written to perform the mathematical calculations required when using Mie theory to aid in determining particle size from Mie scattered light. The MatScat program offers a method to solve for alumina’s Mie coefficients \((a_n, b_n)\), scattering efficiency \(Q_{sca}\), and scattering amplitude functions \(S_1\) and \(S_2\) based on certain material properties of the alumina particle. These values ultimately lead to calculation of the theoretical extinction cross section \(\sigma_{ext}\) and single particle scattering phase function \(p_\theta\).

MatScat plots of the single particle scattering phase function \(p_\theta\) for each alumina particle size used in the experiment are shown in Figures 8 through 11 below.
Figure 8. MatScat generated phase function plot for 0.05 micron alumina particle size

Figure 9. MatScat generated phase function plot for 0.3 micron alumina particle size
Figure 10. MatScat generated phase function plot for 1 micron alumina particle size

Figure 11. MatScat generated phase function plot for 3 micron alumina particle size
It should be noted that the 0.05 micron phase function plot shows a significant amount of light scatter occurring in all angular directions. This type of scattering is characteristic of particles much smaller than the wavelength of incident light and is called Rayleigh scattering. Due to this, Mie theory calculated particle parameters for a 0.05 micron particle size will not be accurate. However, the phase functions for the 0.3, 1, and 3 micron alumina particles show light scatter occurring in the forward angle direction which is characteristic of Mie Scattering.

MatScat calculated values of the theoretical extinction cross section ($\sigma_{ext}$), for each alumina particle size used in the experiment are shown in Table 2 below.

**Table 2.** MatScat generated values of extinction cross section for all different alumina particle sizes.

<table>
<thead>
<tr>
<th>ALUMINA PARTICLE SIZE</th>
<th>EXTINCTION CROSS SECTION ($\sigma_{ext}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05 Micron</td>
<td>0.0018</td>
</tr>
<tr>
<td>0.3 Micron</td>
<td>2.0140</td>
</tr>
<tr>
<td>1 Micron</td>
<td>1.6380</td>
</tr>
<tr>
<td>3 Micron</td>
<td>2.5522</td>
</tr>
</tbody>
</table>
3.7 Using MatScat calculated values to generate theoretical angular distribution function

By using MatScat calculated values for extinction cross section and phase function, a theoretical angular distribution function was calculated for each alumina particle size shown in Figure 12 below.

**Figure 12.** Theoretical angular distribution function calculated for each alumina particle size.
4.0 EXPERIMENTS PERFORMED USING THE FAST TECHNIQUE

4.1 Experimental setup using FAST technique

The setup used for the FAST technique experiments performed at the NCPA was based off of the experimental setup Nefdov used which is outlined in chapter 3.2. The specifications for components to this experimental setup include:

- 20 mW 1135P JDS Uniphase laser
- Spectral Filter (not included in Nefdov setup) (used for “cleaning laser signal”)
- wood rotating disk with aperture diaphragms of diameters 5.0, 7.6, 11.7, 17.3, 26.0, 39.5, 59.5 and 75.0mm
- Thor-labs Plano-convex lens condenser
- BSP = ThorLabs 10% beam-splitter
- GP = 50% glass plate beam-splitter
- \(PD_r (reference) = \) Electro-Optics Technology OT Silicon PIN detector ET-2020 photodiode
- \(PD_a (auxiliary) = \) Electro-Optics Technology Silicon PIN detector ET-2030 photodiode
- \(PD_b (basic) = \) Electro-Optics Technology Silicon PIN detector ET-2030 photodiode
- Tektronix TDS 2024B four channel digital storage oscilloscope
- BNC model 565 pulse delay generator
• National Instruments PXIe-1082 computer

In order for the photodiodes to take light intensity measurements simultaneously and in coordination with the rotating disk, an oscilloscope was connected with a pulse generator. The pulse generator in conjunction with the oscilloscope would send a signal to the computer to take light intensity measurements with a frequency proportional to the angular frequency of the disk. For example, if the disk is spinning at 120 rpm, the pulse frequency will be 0.0625 seconds. At 120 rpm, the disk is spinning at 2 rotations per second, and since there are 8 apertures on the disk, there will need to be light intensity measurements taken at intervals of 0.0625 seconds. The computer program used to run the experimental setup and analyze light intensities after each measurement was written in LabView by Dr. Adam Green.

4.2 FAST technique for alumina in solution

For the purposes of verifying that the FAST setup was functioning properly, experimental trials using the FAST technique for alumina particles suspended in ethanol solution were performed in room 1052 of the NCPA. The experimental FAST setup for these preliminary trials is shown in Figures 13 and 14 below.
Figure 13. FAST Setup in room 1052 of NCPA view 1.

Figure 14. FAST Setup in room 1052 of NCPA view 2.
The particle sizes used for these trials were 0.05 micron and 0.3 micron. Solution preparation was held constant between particle sizes. For each particle size 4 different solution concentrations were prepared and tested shown in Table 3 below.

Table 3. Solution preparation steps for FAST measurements.

<table>
<thead>
<tr>
<th>PARTICLE SIZES:</th>
<th>STEP 1: AMOUNT OF ALUMINA</th>
<th>STEP 2: AMOUNT OF 190 PROOF ETHANOL</th>
<th>STEP 3: AMOUNT OF 0.1M HCL</th>
<th>STEP 4: TIME OF SONICATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05, 0.3 MICRON</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Event 1 (standard used for initialization)</td>
<td>0.0g</td>
<td>50mL</td>
<td>1mL</td>
<td>7 minutes</td>
</tr>
<tr>
<td>Event 2</td>
<td>0.1g</td>
<td>50mL</td>
<td>1mL</td>
<td>7 minutes</td>
</tr>
<tr>
<td>Event 3</td>
<td>0.2g</td>
<td>50mL</td>
<td>1mL</td>
<td>7 minutes</td>
</tr>
<tr>
<td>Event 4</td>
<td>0.4g</td>
<td>50mL</td>
<td>1mL</td>
<td>7 minutes</td>
</tr>
</tbody>
</table>

4.3 Measurements of the FAST technique for alumina in solution

For each particle size tested, measurements were made at a wheel rotation of 120 rpm. Solutions were contained inside a constructed apparatus comprised of \( \frac{1}{8} \) inch thick acrylic sheets held together by an epoxy adhesive. Once light intensities were recorded at each photodiode, the LabView program generated values for the experimental angular distribution function. Plots of the angular distribution function for each event number and particle size are shown below in Table 4. Aperture number go in order from smallest to largest diameter aperture.
Table 4. Angular distribution functions for different solution concentrations of 0.05 and 0.3 micron alumina particles.
The experiments performed using the FAST technique for 0.05 micron and 0.3 micron alumina particles suspended in ethanol solution aided in reassuring the functionality of the experimental setup. However, the data obtained during these trials was not useful for validating the manufacturer labeled alumina size. The reason for this was a combination of the tested solutions being too concentrated (resulting in multiple scattering), along with defects in the tested solution containment apparatus.

These trials did provide useful information on how to improve the future experiments for alumina in flow. Since the alumina would not be contained in an apparatus while in flow, the issue of an apparatus defect would not be relevant. However, evenly dispersing the alumina with enough powder to ensure light scatter but not too much as to promote multiple scattering would pose a significant challenge.

4.4 FAST technique for alumina in high speed flow

The experiment was transitioned from the optics table in 1052 to a mobile setup. The mobile FAST setup was accomplished by fixing each component onto small optical tables each fixed to a single long aluminum rail system hoisted up by two tripods. This allowed for the convenience of moving the intact experimental setup to which ever testing location it needed to be. A picture of the mobile FAST setup is shown in Figure 15 below.
In order for a dispersed cloud of alumina to be introduced properly into the mobile FAST optical setup measurement space, the alumina powder was fed through a constructed PVC piping system.

The system began with a container for inserting alumina powder. From here a small auger, connected to an electronic motor, fed the alumina into a space where pressurized air thrusted the powder through a vortex generator creating an evenly dispersed cloud of alumina particles. The vortex generator was designed using the program FUSION 360 and constructed using 3D printing in the Makers Space of the CME at the University of Mississippi. Pictures of the vortex generator (both assembled and disassembled) along with the PVC piping setup are shown in Figures 16-18 below.
Figure 16. Vortex generator (disassembled).
Figure 17. Vortex generator (assembled).

Figure 18. Combined Vortex generator and PVC piping system setup.
4.5 Measurements of the FAST technique for alumina in high speed flow

Measurements using the mobile FAST setup took place in the anechoic jet lab chamber located in the NCPA shown in Figure 19-20 below.

Figure 19. Mobile FAST setup in Jet Lab of NCPA view 2.

Figure 20. Mobile FAST setup in Jet Lab of NCPA view 3.
For each particle size tested, 5 measurements were made at a wheel rotation of 600 rpm. Once light intensities were recorded at each photodiode, the LabView program generated values for the experimental angular distribution function. Angular distribution values were averaged over the 5 measurements for each aperture. Plots of the averaged angular distribution function for each particle size are shown below in Table 5. Aperture number go in order from smallest to largest diameter aperture.

**Table 5.** Average angular distribution function for all alumina particle sizes subjected to air flow.

<table>
<thead>
<tr>
<th>Aperture number</th>
<th>0.05 Micron</th>
<th>0.3 Micron</th>
<th>1 Micron</th>
<th>3 Micron</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aperture</td>
<td>Aperture</td>
<td>Aperture</td>
<td>Aperture</td>
<td>Aperture</td>
</tr>
<tr>
<td>q(θ)</td>
<td>q(θ)</td>
<td>q(θ)</td>
<td>q(θ)</td>
<td>q(θ)</td>
</tr>
</tbody>
</table>

Diagram showing plots of q(θ) for each aperture size.
5.0 CONCLUSION

The LS spectrometer was able to validate manufacturer labeled 0.05 micron, 0.3 micron, 1 micron, and 3 micron alumina particle sizes while suspended in ethanol solution using a single angle measurement. Successful solution preparation required a very small particle mass ( < 0.01g ) of alumina in order to limit the effects of multiple scattering.

Experiments performed using the FAST technique were ultimately not successful in validating alumina particle size under both environmental conditions. The results obtained undertaking this effort underscore the complexity of attempting to control all the different variables in this experiment. However, the FAST technique should not be looked upon as a flawed method for obtaining particle size. Particular problems noted throughout this experimental process certainly contributed to invalid data acquisition.

These include:

- Use of linearly polarized laser light
- Photodetectors not sensitive enough
- Unalignment and warping of wood disk resulting from repeated high rpm measurements
- Epoxy adhesive effecting transparency of acrylic containment apparatus (for solution measurements)
- Inability to introduce a consistent, and appropriately dispersed amount of particle mass into the sample space (for air flow measurements)
• The 0.05 micron alumina particle size parameter being too small resulting in the Rayleigh scatter of light.

Solutions to these problems include the implementation of a laser depolarizer. Since calculating the phase function for the theoretical angular distribution function requires the assumption of unpolarized light, it is important to implement a laser depolarizer into the experimental setup.

Also, adding more sensitive photodetectors into the setup would allow for better detection of scattered light from experimental measurements.

Additionally, removal of the rotating disk and instead applying a motorized iris shutter would negate previous issues of warping and unalignment due to high rpm measurements. Utilizing an iris shutter would also provide a smoother angular distribution function as forward scattered light could be collected through a more numerous spectrum of aperture diameters.

For solution measurements, constructing a solution containment apparatus using an adhesive alternative, such as 3M clear adhesive, should alleviate the negative effects epoxy had on transparency.

For air flow measurements, Nefedov offers a technique to introducing a steady concentration of particle mass into a sample space utilizing a laminar-diffusion-flame design (Nefedov, 1997). For this design, a flat flame burner is composed of multiple flames in a closely spaced array. These flames are fueled through the combustion of propane with air. Flow rates for both propane and air are controlled by calibrated flowmeters. A fluidized bed of particles is admixed with the air to inject the particles into the body of the flame generating a dispersed aerosol.
Successful application of the FAST technique depends on the Forward scattering of light. Therefore, implementing a laser with a lower wavelength would increase the size parameter for the 0.05 micron alumina particle size and promote the Mie (forward) scattering of light.

Due to time constraint, solutions to the noted problems above were not able to be applied into the experimental setup. However, if implemented for future measurements, these discussed solutions should improve acquiring valid data using the FAST technique.


[17] Schäfer, J. and Lee, S.-C. and Kienle, A., Calculation of the near fields for the scattering of electromagnetic waves by multiple infinite