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Final Report

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Determination of Particle Size Using Measurement of Scatter

To

National Aeronautics and Space Administration

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Summary

A literature search was conducted to determine the state of the art for particle size measurement by the light scatter technique. This technique may involve diffraction pattern analysis, location of minima and maxima in angular dependence of scattered light, magnitude of intensity verses angle, forward lobe scattered intensity ratio using two small angles, forward scatter in a small cone, and total scatter. Some of the more modern recordings and detection systems are video, holographic, and systems using optical processing.

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Particle Sizing By Scattering

INTRODUCTION

Sizing particles by scattering falls into several categories which depends upon the nature of illumination, particle size, number of particles illuminated and concentration.

The illumination may be a single wavelength as with a laser, a wave band, or some sort of lamp. Also, the light source may be coherent or noncoherent. The illumination-detection system may be arranged such that only a single particle is seen at a given time. This gives the measuring system counting capability. The specimen illuminated may be a volume of particles of the same diameter (monodisperse) or it may be particles of different diameters (polydisperse). In both cases low concentrations can be analyzed by single particle theory. For high concentrations the system of particles may need to be diluted before analysis. This can introduce error if the particles can evaporate or condense. Errors can be kept to a minimum if the diluting gas is of proper temperature, pressure, and humidity.

Particle sizing by scattering include diffraction pattern analysis, location of minima and maxima in angular dependence of scattered light, magnitude of intensity verses angle, forward lobe scattered intensity ratio using two small angles, forward scatter in a small cone, and total scatter. The experimental system for using the above traditionally consists of a source, detector, and perhaps some device for changing the angular position of the detector. In some instances a microscope and a camera can be used.

DISCUSSION OF LITERATURE

Cornillault (11) has discussed a method of measuring particle size distribution using diffraction pattern analysis. He uses a lens to focus the diffraction pattern from the system of particle in the plane of the detector. The detector is moved perpendicular to the axis of the len. According to Cornillault, the size distribution can be linearly related to the detector measurements and he discusses how these measurements are related to Bessel functions. Sets of data and calculation of size are not given. However, he indicates the technique is valid for particles whose diameter is four times greater than the light wavelength.

Belz (5) has a fairly thorough analysis of the use of diffraction for particle size analysis. His work includes a derivation and discussion of the use of intensity variations in the diffraction pattern of a single particle to size particles. Cinsiderable discussion is also given on the reconstruction of the hologram obtained from the diffraction pattern. Numerous figures obtained from the theoretical and experimental results are given. Considerable data are given that would aid one in beginning an experimental program in particle sizing. Also, many references are cited that would be helpful for further study in this area.

Kerker, et al, (21) has described a technique of measuring particle size by measuring the location of the maxima and minima in the scattered light. The measurements begin from the forward direction. Kerker was concerned with the validity of the equation

$$\left(\frac{D}{\lambda}\right)_m \text{Sin}\left(\frac{\Theta_i}{2}\right) = \begin{cases} k_i \\ K_i \end{cases}$$

for use in this technique. D is the particle diameter, λ is the light wave length, Θ_i is the angle at which the maximum or minimum occurs and k_i and K_i are parameters corresponding to the ith minimum and ith maximum. This equation is a very simple means of obtaining D from values of λ, Θ_i, k_i and/or K_i. Using Mie solutions Kerker, et al, computed values of k_i and K_i and drew 2% and 10% error contours. They pointed out in their discussion that it may be difficult to assign the proper index to the various minima and maxima because the detection system may not be able to obtain valid measurement near 0^o. This is a primary concern for error in this method.

A graph of intensity verses angle can be used to measure size. For a single particle one measures the variation of intensity with angle and compare this with a theoretical result or calibration curves. Aerosols of high concentrations may have to be diluted and/or the incident light may have to be focused or collimated so that only one particle intercepts the beam at a given time. Marshall, et al, (23) have reported 360^o scattering diagrams for a single particle (polytyrene spheres). From these diagrams they indicate diameter and index of refraction can be determined to an accuracy of about 0.4 percent and 0.7 percent respectively by using a computer to compare their data with theoretical Mie solutions. They believe their method is valid for diameters between 0.3 microns and 20 microns. In addition to the above, Marshall, et al, discussed varing both index of refraction and size and comparing the results to theoretical curves. This allowed for the determination of the index of refraction. Kerker (21) has also discussed this technique of sizing particles. He discusses the difficulty in using this method when more than one particle is illuminated at a given time.

Essentially, the scatter from the various particles add in a way that makes it impossible to identify the particle size by simple comparison with Mie solutions.

Ivlev, et al, (19), Boron, et al, (7) and Gravatt (15) have discussed the technique of sizing a particle that utilizes the ratio of intensity computed from measurements at two small angles near the forward direction. The primary advantage of this method is that the results are not dependent on the index of refraction. According to Gravatt, who illuminated a single particle using this method, the sizing error is not greater than ± 15% for a range in size from 0.2 to 4 microns. He also indicates the method may be extended to the 0.05 to 5 micron size range. Angles between 2.50 to 20° are used to obtain the data for a 500 nm source. In addition Gravatt discusses a technique of determining the chemical composition which used the intensity integrated over 40° to 70°. Boron's, et al, paper is a discussion of the theoretical errors one would have when using this method. The errors are due to the assumption that the intensity ratio is not a function of the index of refraction which is not strictly true. It should also be pointed out that this method does not require a uniform light beam as with most methods. This is because a ratio of intensities are used instead of an absolute intensity.

Another technique which uses a light scattering measurement system is that of forward scatter, Mikirov (24). Mikirov used a parallel beam of light to illuminate the specimen. When there is no particle in the light the beam is cut off by a dark screen and no light reaches the photo cathode of a photo-multiplier. According to Mikirov 86.4% of the light incident on the particle will be used when the photo-multiplier intercepts the light which goes into the forward π steradions. Mikirov's illuminated volume is chosen so that there is a 0.05 probability that not more than one particle is in the light beam. He gives data for cloud particles in the 2 to 30 micron range.

Schulz, et al, (31) use forward scatter in their design of a light scattering cascade impactor. The light scattering system was developed in the forties by Knulson and White and is described in NRL Report p-2642. The interesting aspect of their design is the 45° illumination of the particle. A lens is used to focus light on the particle but the lens is covered with tape except for its outer edge. Using the lens with proper focal length, they made the source imping upon the particle at 45°. The photo-tube was placed on a line passing through the particle, lens, and source. This eliminated the component of source that normally hits the photo-tube directly.

A novel forward scattering technique is that by Weiss and Frock (35). They use a laser to illuminate the particle and this causes the typical bright and dark rings which depend on the particle size. The novel aspect of the technique is the use of a spatial filter between the particle and photodiode to transform the intensity of the diffracted light such that the photodiode responds to the particle volume and diameter. The photodiode signal is processed with a MCS-8 micron processor and the results are displayed on a digital LED array or a printer gives a hard copy.

A similar technique is that of measuring the total scatter from a single particle. Jacobi, et al, (20) has discussed such a measurement system. The system uses a laser while that of Mikirov's used a conventional light source. Jacobi's system was calibrated with latex spheres for the 0.5 to 2.5 micron region. According to Jacobi, his calibration curve can be corrected for other index of refractions for spheres in the 0.5 to 2.0 micron range. He chooses his illumination volume so that there is less than a 5% coincidence probability.

Another method of determining particle size is to obtain measurements of scatter from a system of particles and invert the appropriate equation to obtain the particle distribution. Most of these equations require some assumptions on the nature of the size distribution. Unlike most of the other techniques, this method has as it's primary difficulty the mathematical inversion of a transmission or scatter equation. Contributors to this method include Shifrin and Raskin (30), Twomey and Harrell (34), Phelps (27), Abbiss (1), Cohen (9), Anderson and Bessiner (4), Herman, et al, (16), Westwater and Cohen (36), and Post (28). These authors are primarily concerned with the analytical details of inversion, quantity and accuracy of experimental data needed for dependable reproduction of the size distribution. A successful inversion technique has the advantage of requiring a fairly simple measurement system. But because the result is usually a curve, this method may be erroneous when the particle sizes are at few discrete intervals.

Several investigators have used the laser scatter from particles to produce a hologram. Kunkel (22) used a hologram camera that was designed by Silverman, et al, (32) to measure the size and concentration of fog droplets. He reports measurements of drops between 4 microns to 70 microns. For 4 microns his sample volume was $0.8 cm^3$ and $4.5 cm^3$ for sizes greater than 40 microns. His paper does not discuss the theoretical aspects of holographic design since he uses an existing design with few modifications. Stigliani, et al, (33) gives a theoretical and experimental discussion of the use of Fresnel Holography to measure the size of glass spheres. Their measurements are for 80 to 250 microns using a 632.8nm helium-neon laser. The spheres were suspended by using a 14 micron silk thread. Experimental and theoretical results are presented as graphs of normalized radiance as a function of distance along the sphere's radial axis. The experimental radiance distribution was obtained from the photograph with a densitometer. Their experimental data was within 4% of their theoretical results.

One of the best publications on the details of experimental holography is that of Belz (6). His publication gives many of the details one needs to begin investigations in holography. Some of the topics which are discussed in this paper are particle size limits, recording distance, depth of field, out-of-focus images and magnification of particle field. In many cases this author discusses the optimal value of the experimental variables. He gives data for spray paint for sizes above 25 microns.

The holographic technique has been used by several investigators to study specimens that have high velocities. Pavitt, et al, (3) have investigated the holographic sizing of particles in the 15-1000 micron range using the Gabor in-line technique. He used a Q-switched ruby laser for construction of the hologram and a helium-neon laser for reconstruction. The velocity of the particle was approximately 100m/s. He discusses the coherence requirements, photographic emulsion requirement and effect of the particle motion on holograms.

Troiinger, et al, (6) have investigated the holographic recording of the dynamics of a particle field. They produced double exposed in-line holograms taken near a high pressure atomizer. The diameter of the droplets (water) were between 20 and 150 microns and the velocities varied up to 15m/sec.

Another excellent publication on holography is that of Zinky (38). Even though this publication is primarily concerned with measuring particle size in explosively generated aerosols, many topics are discussed that are of interest to one beginning in this field. The publication describes the design of a hologram camera and reconstruction system for the 3 to 1000 micron range. Theoretical derivations are not given but important results from theory needed to define system and component requirement are discussed. These include resolution requirements of the film, effects of spatial and temporal coherence on resolution, film processing and alignment of the system.

Itagaki (18) have used the microscope to measure water droplets in the 5 to 45 micron range. The primary disadvantage of this method is the small (3×10^{-4} ml for 30 X Magnification) sample volume. This is partially overcome by using multiple exposures on a 34 mm film.

Cole, III, et al. (10) have presented an optical method based on measurements of transmission vs time at two wavelengths to size particles in a cloud chamber. The measurements were interpreted using the Mie scattering theory. The interpretation technique is to compare the analytical details of the experimental curves with theoretical curves. Final droplet diameters from 1.75-10.4 micron with corresponding concentration from 1.8×10^{-6} -1.15×10^{4} cm^3 were measured.

Hotham (17) gave a presentation on the use of a particle sizing system which incorporates a laser, TV camera, monitor and video recorder. The system measure particle sizes from 0.3 to 1000 micrometers. His discussion is primarily concerned with explaining several photographs that was obtained with his system (which appear to be of a high quality).

Gollub, et al, (13) and Yanta (37) have used doppler shift caused by the velocity of the particles to determine particle size. Both methods require that the velocity be known. Gollub's work was for a cloud chamber in which the settling velocity is given by Stokes Law. Yanta placed his particles in a supersonic flow for which a procedure for computing the particle velocity had been developed. His comparison of his results with optical o w 1 showed very good agreement.

CONCLUSION

First I should mention a report by Ranade (29). This report is unlike th. previously discussed papers in that it is concerned primarily with systems for assessing aerosol. Many off the shelf systems are discussed. Typical information included are principle of operating, size range, accuracy and cost. One should study this report before purchasing a particle measurement system.

This points to the fact that the field of particle size measurement is a fairly well developed one. Research problems in this field are primarily due to special requirements on accuracy, space and perhaps size range.

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