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**Development of Laboratory and Process Sensors to Monitor
Particle Size Distribution of Industrial Slurries**

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ABSTRACT

In this paper we present a novel measurement technique for monitoring particle size distributions of industrial colloidal slurries based on ultrasonic spectroscopy and mathematical deconvolution. An on-line sensor prototype has been developed and tested extensively in laboratory and production settings using mineral pigment slurries. Evaluation to date shows that the sensor is capable of providing particle size distributions, without any assumptions regarding their functional form, over diameters ranging from 0.1 to 100 micrometers in slurries with particle concentrations of 10 to 50 volume percents. The newly developed on-line sensor allows one to obtain particle size distributions of commonly encountered inorganic pigment slurries under industrial processing conditions without dilution.

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INTRODUCTION

Slurries, mixtures of finely divided solids dispersed in liquids, are found throughout the industrial sector, but occur most commonly in the chemicals, food processing, and pulp and paper industries. In the chemicals industry alone, the production of thousands of intermediate and final products involves the use of slurries. The size of solid particles in a slurry directly affects the characteristics and quality of the final product, and the energy requirements of subsequent processing steps. The present lack of on-line particle size distribution information is a major cause of poor grinding control and low yields in several slurry processing operations. Under grinding leads to poor quality product. Over grinding leads to excessive energy consumption. Applications of on-line particle size distribution sensors would help avoid under or over grinding.

There are several methods available for measuring particle size distributions in slurries, but many of them have significant technical limitations in the context of on-line monitoring. Presently, there is no suitable sensor available to monitor particle size distributions in slurries containing sub micron particles at high solids loading. Current methods require the preparation of diluted samples in a laboratory, a time consuming procedure that renders these methods of limited use in process control applications. Methods based on sizing individual particles using electrical sensing zone technique, light-blocking or light-scattering techniques are usually applied to dilute systems.

Recently, a new on-line sensor was developed for monitoring particle size distributions of industrial colloidal slurries. The sensor is based on estimation of particle size distribution functions using measurements of ultrasound attenuation coefficients at several frequencies by solving appropriate integral equations. The mathematical algorithm employed in the deconvolution or inversion of measured spectra utilizes the results of a recent theoretical model developed to predict the behavior of ultrasound as it

passes through a slurry of polydisperse particles at a known concentration, over the range of 1 to 50 volume percent.

During this project, Pen Kem developed and tested a prototype sensor electronics and computer system with the measurement software capable of programmed ultrasonic spectroscopy. The slurry characterization chambers and the sample delivery system along with the sensor operating software were developed by P&G Systems with design and fabrication assistance from Techna Machine. The new sensor system was tested successfully in the University of Maine and Du Pont research laboratories. An on-line prototype was evaluated at Du Pont's semiworks facility for pigment production during several months of field testing.

Testing and evaluation to date has shown that the new ultrasound based particle size distribution sensor system is capable of handling several types of mineral pigment slurries commonly encountered in industry under processing concentrations. During the field tests, the system has operated reliably as an on-line monitoring tool during protracted runs of continuous operation extending over several days, providing data sensitive to changes in the processing conditions. Further work is underway to adapt the ultrasonic spectroscopy for shape characterization of particles in suspensions.

DESCRIPTION OF THE NEW ACOUSTIC PARTICLE ANALYZER

The acoustic particle analyzer relies on the accurate measurement of attenuation coefficients of suspensions, for sound frequencies ranging from 3 to 100 MHz, using a rugged, flow-through Slurry Characterization Chamber (SCC) equipped with a pair of broad-band ultrasonic transmitter and receiver transducers. The transducers are mounted to provide real-time, programmable, variable gap capability with less than 1 μm resolution while maintaining parallelism between the transducer faces. A custom

designed computer controlled electric signal processing hardware provides for generation of suitable tone-burst signals to be applied to the transmitting transducer, and detection of amplitude¹ of the signals from the receiver transducer set at selected gaps for selected frequencies while taking appropriate care in time-domain rejection of spurious signals. After obtaining the measurements of signal loss (as high as 140 dB) corresponding to several gaps and frequencies for all selected chambers the attenuation coefficient² spectra are computed for each chamber based on screening of individual data points using measured noise levels and estimations of spurious contributions for certain combinations of gaps and frequencies. The attenuation spectra along with the corresponding estimates of quality factors obtained from each chamber are then used to compute a composite spectrum. These attenuation spectra then are used by the deconvolution software to estimate corresponding particle size distributions³. In a laboratory system, a slurry characterization chamber is integrated with a circulating pump, as well as temperature, pH and conductivity probes. In a on-line system, a slurry characterization chamber

¹ In a proprietary implementation both amplitude and phase data is obtained.

² In a proprietary implementation sound speed spectra over the entire frequency range are also obtained by analyzing the phase data.

³ In a proprietary implementation the sound speed measurements are used to compute the total particle concentration. This concentration estimate is used in the deconvolution of attenuation coefficient spectra to obtain particle size distributions. This feature is important since it combines the measurements of sound speed and attenuation coefficients to get the particle size and concentration information. The technologies based on prior art do not obtain and use sound speed information. Earlier attempts to obtain both particle size distribution and concentration from the attenuation coefficient spectra have led to several difficulties and limitations.

assembly (SCCA) consisting of two/three chambers and two temperature sensing modules is mounted in NEMA-4 enclosure. It is integrated with a computer-controlled sample delivery system (SDS) capable of handling a continuous bypass slurry stream from an industrial processing unit, and a supply of fresh water for programmed washing of certain sections of the slurry characterization chamber assembly. The SDS is designed to provide capability of programmed interruption and resumption of the measurement sequence to flush the critical slurry path at suitable intervals. The SCCA is designed to allow for easy field service/replacement of the chambers or chamber components. A schematic diagram of a on-line system is shown in Figure 1.

Fig. 1. Schematic diagram of the AcoustoPhor on-line prototype for determining particle size distribution of industrial colloids (SCCA = Slurry Characterization Chamber Assembly; SDS = Sample Delivery System; SOC w/SE = Sensor Operating Computer with Sensor Electronics)

While a section of the electric signal processing hardware is mounted in the SCCA enclosure in a close proximity to the chambers, the operating computer and the associated hardware in the computer cabinet can be set in a convenient place, such as a control room on the mill floor, several hundred feet away from a processing unit. The operating computer is also equipped with a provision for data-transfer to a mill-computer, in real-time, for ease of integration of these particle size distributions in process control. A working prototype of a on-line system has been tested at a pigment manufacturing site at a Du Pont plant for several months in early 1992.

TEST DATA AND PERFORMANCE EVALUATION

General Issues related to Particle Size Analyzer Evaluation

Several measurement techniques for obtaining particle size distributions of slurries with on-line or rapid off-line sampling have emerged lately as discussed by Terry Allen in his book entitled "Particle Size Measurement" (Fourth edition, 1990). Several of these are particle counters that involve analysis of individual particles using optical or electric sensing zone techniques, and hence are restricted to very dilute systems often encountered in pollution or contamination control applications. Techniques involving analysis of an assembly of particles often use laser diffraction pattern analysis allowing analysis of slurries at moderate concentrations. Low-angle laser light scattering (LALLS), Fraunhofer diffraction, analysis is widely used in several commercial instruments which are now being used for rapid off-line and on-line use mostly when particle larger than one micron are involved. For particle size range of 0.005 to 5.0 microns, photon correlation spectroscopy based commercial instruments have been widely used for rapid off-line analysis. All of these instruments usually require dilution of process slurries. A notable exception is that of Lasentec's scanning laser back-scattering microscope probe, which

has been reported to work on slurries, with concentrations as high as 20 volume percent, when particles are larger than few microns. One ultrasound-based instrument has been used mining industry on concentrated slurries when particles are larger than 40 microns. *Our contacts with several companies involved in slurry processing (for example, Du Pont Chemicals, Engelhard, Pfizer, and Alcoa), especially with the process engineers in these companies, have met with substantial interest in the new ultrasound-based particle size distribution analyzer capable of handling slurries of submicron particles under processing concentrations that are often 10 to 50 volume percent.*

In the particular context of inorganic pigment slurries encountered in industrial processing, the laboratory instruments using x-ray sedimentation analysis (for example, SediGraph 5100), which can not properly account for particles smaller than 0.2 microns, appear to most common in industrial product quality control laboratories. The process slurry sample is usually transported to an on-site laboratory, if available, where it is diluted and analyzed using a laboratory instrument equipped with an automated multiple sample handling system. Individual analysis often takes 30-45 minutes. Recently, x-ray disc centrifuge (BI-XDC), which can sense particle smaller than 0.2 micron, has become available but it still is not widely used. The XDC also requires the dilution step, but is faster with individual analysis often taking only 8-10 minutes. Data from such laboratory instruments however can not be used in process control applications mainly because of long delays associated with process slurry analysis resulting from sample handling details. *There are currently no commercial instruments available to analyze industrial process slurries selected for analysis in this study (for example 75 wt% Titanium Dioxide, 55wt% Kaolin clay, or 65 wt% Calcium Carbonate) as received. In the present study, our ultrasound-based particle size distribution data was compared with the SediGraph data for comparisons, in spite of the limitations of the SediGraph technique. The*

conventional industrial wisdom regarding process or product quality control is based on accumulated experience on SediGraph data analysis, and most process engineers like see any new technique offer results that correlate with SediGraph analysis. Some users may want to compare results to Fraunhofer diffraction based instrument such as Leeds and Northrup Microtrac or Malvern Mastersizer. This is an important aspect of introducing a novel measurement technique for particle size distribution analysis to the slurry processing industry.

In slurries particles often exist as aggregates (firmly held clusters) or agglomerates (loosely held clusters) depending on the state of dispersion. When one talks of particle size distributions some refer to size distribution of individual particles if all agglomerates were broken by dispersing them well as is often done by dilution and addition of a dispersant. On the other hand, some refer to size distribution of agglomerates as they exist in the sensing zone under a given state of dispersion. This causes differing expectations by users from particle size analyzers. *In the on-line process control context, potential users are looking for particle distribution data on their process streams in real-time under processing conditions, (within 1 to 10 minutes!), that would correlate to selected end-use tests and that would be responsive to changes in selected processing variables.* Such rapid response particle size distribution data should reflect true changes in particle size distributions, and not changes in concentration, flow rate, or temperature of the slurry.

Test Slurries

The following slurries were used in this study to evaluate performance of the newly developed sensor prototype. These samples were selected on the basis of our estimates of related energy savings potentials, and identification of specific technically and economically justified on-line control opportunities. Two types of slurries were

involved: (1) Finished products which are often stable, well dispersed, and easy to handle; and (2) In-process samples, which are often unstable, flocculated, and more difficult to handle. Slurries were kindly supplied by the manufacturers with appropriate data on particle size distribution from the SediGraph, particle density and particle solids level.

The test slurries involved three papermaking pigments:

1. Titanium Dioxide (produced by gas phase powder synthesis),
2. Kaolin Clay (produced by mining),
3. Calcium carbonate (produced by precipitation reaction).

Most of the testing and evaluation was performed at the University of Maine. Laboratory tests involved analyses of slurries as received, slurries treated with different mixing conditions, slurries prepared by dilution with water, and slurries modified by addition of flocculants or dispersants. For the particular case of titanium dioxide pigment slurries, we worked at the Manufacturing Development Unit at Du Pont's New Johnsonville, Tennessee plant. The plant tests focused on robustness of the prototype, repeatability of data under controlled conditions, and sensitivity to changes in selected operating parameters on a grinding unit in several protracted runs.

Overall Performance Evaluation⁴

During the initial stage of the project, in June 1990, high solids slurries of Titanium Dioxide were used for feasibility analysis. In a 'blind test' three 70 weight % samples,

⁴ All the test data and details of analysis will be completely documented in Sharma's Ph. D. dissertation. Theoretical model has already been documented in Strout's Ph. D. 1991 dissertation. Several technical publications are expected in the near future. General theoretical background is given in the Appendix.

supplied by Du Pont Chemicals were analyzed using a laboratory acoustic spectrometer setup. Acoustic technique was able to identify one commercial sample distinguishing it from the other two experimental samples. Furthermore, it showed ability to correctly identify the ground slurry from the unprocessed slurry. Tests were repeated one week later. Data were found to be fairly reproducible. These data were reviewed by the Du Pont staff and the DOE program manager.

At a later stage, in January 1991, additional tests were conducted by Du Pont personnel at the Engineering Test Center (ETC) using the laboratory prototype under the direction of Dr. Terry Allen. Test data were compared with results from the analysis of diluted samples using Sedigraph 5100 (Micromeritics) and BI-XDC (Brookehaven Instruments). These data were reviewed by Du Pont staff in March 1991 with the DOE program manager.

During March 1992 to June 1992, the on-line prototype was evaluated at the Manufacturing Test Unit at Du Pont's New Johnsonville, TN plant. During several twenty-four hours per day, five days per week operations the analyzer was found to operating in a robust and reliable manner. Response time was 10 minutes. The analyzer was found to be sensitive changes in processing variables.

In September 1992 samples of precipitated calcium carbonate (PCC) were analyzed under the supervision of Dr. Nigel Sanders, Senior Research Chemist from Specialty Minerals. Specialty Minerals, a division of Pfizer Inc., is the largest supplier of PCC in the world. Test slurries were at 70 wt% solids. Acoustic analyzer was successful in measuring inter-sample differences in mean size and also distribution width.

APPENDIX: THEORETICAL BACKGROUND

Acoustophoresis and associated Sound Attenuation

When a sound wave travels through a suspension, the suspended particles as well as the suspending medium undergo oscillatory motion. The motion of particles relative to the surrounding medium under the influence of acoustic field is termed "acoustophoresis". Viscous dissipation associated with the acoustophoresis⁵ leads to attenuation of sound. The extent of attenuation depends on the length of the acoustic path, the concentration and sizes of the particles, the frequency of sound, and the physical properties of the particulate and fluid phases.

In particular, the density contrast, σ defined as the difference between the densities of the particulate and fluid phases divided by the fluid density $[(\rho_p - \rho_o)/\rho_o]$ is an important characteristic parameter in connection with the acoustophoresis and hence the viscous attenuation. The acoustophoretic velocity is zero when the density contrast is zero. The expression for the acoustophoretic velocity, v_A normalized by the undisturbed fluid velocity, u takes the form:

$$v_A / u = (4s^2\sigma/9) \cdot C_V(s, \phi) \quad (1)$$

with

$$s = (D^2\omega\rho_o / 8\eta_o)^{0.5} \quad (2)$$

⁵ When suspended charged particles and their diffuse ionic atmospheres are involved macroscopic induced electric potentials are associated with the acoustophoresis. Measurement of the induced electric potentials resulting from the acoustophoresis of colloids has been used for a commercial sensor system (Pen Kem Acoustophoretic Titrator) for monitoring surface charge or zeta potential on colloids.

where

D = particle diameter,

ω = sound frequency in radians/second, $(2\pi f)$,

f = sound frequency in cycles/seconds,

ρ_o = density of the suspending medium,

η_o = viscosity of the suspending medium,

s^2 = vibrational Reynold's number,

σ = density contrast $[(\rho_p - \rho_o)/\rho_o]$,

ϕ = volume fraction of the suspended particles, and

$C_v(s, \phi)$ = acoustophoretic velocity correction factor

Based on the considerations of the rate of viscous energy dissipation in a suspension of spherical solid particles subjected to a sound wave one obtains the following expression for the attenuation coefficient contribution due to monodisperse particles, α_{mono} :

$$(\alpha_{\text{mono}} c_o / \omega \phi) = (2s^2 \sigma^2 / 9) \cdot C_A(s, \phi) \quad (3)$$

where

c_o = sound speed in the suspending medium, and

$C_A(s, \phi)$ = attenuation coefficient correction factor

For the case of polydisperse particles the attenuation coefficient contribution as a function of operating sound frequency f , $\alpha(f)$ becomes:

$$\alpha(f) = \int \alpha_{\text{mono}}(f, D) g(D) dD \quad (4)$$

where

$g(D)$ = particle size distribution by mass fraction with $g(D)dD$ equal to the mass fraction of particles having diameters in range of D to $D+dD$, and

$\alpha_{\text{mono}}(f,D)$ = attenuation coefficient contribution at a frequency f due to a monodisperse population with all particles having diameter equal to D .

The particular expression for α_{mono} mentioned above takes into consideration the viscous dissipation associated with the acoustophoresis properly accounted for finite values of both the vibrational Reynold's number and the particle volume fraction. The non-linearity associated with the concentration dependence of attenuation coefficients is accounted for in the factor $C_A(s,\phi)$. For very dilute suspensions, ($\phi \rightarrow 0$) and for small diameters and low frequencies ($s \rightarrow 0$), $C_V(s,\phi)$ and $C_A(s,\phi)$ approach 1. For suspensions of submicron particles with considerable density contrast the viscous contribution to the attenuation often dominates over the thermal and scattering contributions in the frequency range of practical interest corresponding to the long-wavelength limit ($\lambda \gg \pi D$ and $\sigma \gg 1$). These conditions correspond to ultrafine inorganic pigment slurries commonly encountered in industry.

Accounting for Other Contributions to Sound Attenuation

In general, one can use suitably modified expressions for α_{mono} to take into consideration the viscous, thermal, scattering, and diffraction contributions as warranted. Such expressions can be obtained using the well known analysis by Allegra and Hawley of the scattered wave potential⁶ for a compressional plane wave impinging on an isolated

⁶ In order to use this formulation properly one needs to know several physical parameters characterizing the particle phase at the operating temperature, such as density, thermal conductivity, thermal dialation coefficient, specific heat, and bulk and rigidity moduli. Similarly one needs to know several physical parameters for the suspending fluid medium at the operating temperature, such as density, shear viscosity, bulk viscosity, thermal conductivity, thermal dialation coefficient, specific heat, and bulk and rigidity moduli.

single spherical particle suspended in a viscous fluid. These results however are limited to very low particle volume fractions (less than 5%). Various multiple-scattering extensions of these results using Waterman and Truell theory allow one to obtain expressions applicable to particle volume fractions up to 50% when viscous losses are not significant. It should be noted that the theoretical expressions for α_{mono} are usually quite complicated and their implementation often involves numerical solution of simultaneous algebraic equations with complex (real and imaginary) coefficients and unknowns. The coefficients may involve special mathematical functions with complex arguments. Recently, Pendse and Strout have developed an oscillatory cell model to account for interparticle hydrodynamic interactions. They applied the oscillatory cell model results to describe the viscous coupling encountered in sound propagation through suspensions with particle volume fractions ranging from 1 to 50%. This model is applicable for a wide range of particle diameters, operating frequencies, and particle concentrations but it only accounts for viscous dissipation contributions as mentioned above. A judicious use of several theoretical results allows one to obtain a general relationship between ultrasound attenuation spectra and particle size distributions applicable for commonly encountered industrial colloids.

General Formulation of the Integral Problem for Estimating Particle Size Distribution using Ultrasound Attenuation Spectra

Using measurements of the attenuation coefficient as a function of operating frequency, $\alpha(f)$ covering an appropriate frequency range and a known expression for $\alpha_{\text{mono}}(f,D)$, one can obtain the particle size distribution function, $g(D)$ by inverting the

above mentioned integral equation⁷. The problem encountered in determining $g(D)$ from known $\alpha(f)$ is known by mathematicians as a Fredholm integral problem of the first kind. It is important to reformulate the governing integral equation in terms transformed variables as follows:

$$R(f) = \int K(f,X)p(X)dX \quad (5)$$

where

$$R(f) = \alpha(f) / r(f) \quad (6)$$

$$X = q(D) \quad (7)$$

$$p(x)dX = g(D)dD \quad (8)$$

and

$$K(f,X) = \alpha_{\text{mono}}(f, D(X)) / r(f) \quad (9)$$

The transformations defined by Equations (6) and (7) are selected to obtain appropriately normalized and non-dimensionalized variables R and X . Proper selection of functions $r(f)$ and $q(D)$ can yield a well-behaved Fredholm integral problem. Note that $r(f)$ is independent of diameter D , and $q(D)$ is independent of frequency f . In this context the function $R(f)$ is referred to as the response function, and the function $K(f, X)$ as the kernel function.

In practice the measured response is not known accurately. Hence the above equation is only satisfied within measurement errors. This integral problem does not possess a

⁷ This general approach has been used by several researchers to estimate particle size distribution functions. One specific example involves use of the fractional penetration through an aerosol classifier at different flow rates as the kernel function. (Yu and Gentry, Powder Technology, pp. 79-89, 1987).

solution if the kernel function $K(f, X)$ is separable into factors as $K'(f) \cdot K''(X)$. Hence, for a separable kernel function one can not determine $p(X)$ from known $R(f)$. Another peculiar nature of this integral problem can be appreciated by noting that

$$R(f) = \int K(f, X) \cdot \{p(X) + Q \cdot \sin(\kappa X)\} dX \quad (10)$$

as $\kappa \rightarrow \infty$ for an arbitrary constant Q

for any $K(f, X)$ as long as it is reasonably smooth and not a sharply peaked kernel. Therefore, a true solution $p(X)$ and a test function obtained by an addition of an highly oscillatory error function $\{p(X) + Q \cdot \sin(\kappa X)\}$ both will satisfy the governing integral equation. Thus a Fredholm integral problem of the first kind becomes a non-trivial problem. The success in inverting the Fredholm integral equation of the first kind depends on the measurement errors and the shape of the kernel function.

Representation of the Integral Problem in Matrix Equation Form

The integral equation can be transformed to a matrix equation by first considering discrete values of the operation variable, (frequency f) and then replacing the integral by an appropriate quadrature formula using a weighted sum of the base-point functional values of the integrands as shown below:

$$R_i = \int K_i(X) p(X) dX \text{ for } i = 1, 2, \dots, M \quad (11)$$

$$K_i(X) = K(f_i, X) \text{ for } i = 1, 2, \dots, M \quad (12)$$

$$R_i = \sum_{j=1}^N K_{ij} W_j p_j \text{ with summation over } j = 1, 2, \dots, N; \\ \text{for } i = 1, 2, \dots, M \quad (13)$$

where

W_j = Quadrature weights for the numerical integration formulation,

$K_{ij} = K(f_i, X_j)$

$R_i = R(f_i)$, and

$p_j = p(X_j)$

Defining

$$S_j = W_j p_j \quad (14)$$

we get

$$R = K \cdot S$$

where $R_i = \sum_{(j)} K_{ij} S_j$ with summation over $j = 1, 2, \dots, N$;

for $i = 1, 2, \dots, M$ (15)

Here, K is an $M \times N$ matrix, and R and S are column vectors of dimension M and N respectively. One needs to pay attention to problems associated with near-singular coefficient matrix, K involved in the above equation. There are several algorithms reported in literature to solve Fredholm integral problems of the first kind. However, no single algorithm is found to be suitable for handling several widely different kernel functions when the response function is known with only modest accuracy as commonly encountered in practice.

We have extensively studied several deconvolution algorithms⁸ using simulated response functions appropriate for ultrasonic spectroscopy of industrial colloidal slurries. We have been quite successful in working with unimodal distributions, as well as bimodal distributions. In terms of its ability to differentiate two distributions with

⁸ The deconvolution algorithms include the following: Non-linear Iteration, Constrained Linear Inversion, Singular Value Decomposition, Cross Validation Inversion, and their various combinations.

relatively small differences in means and standard deviations our present deconvolution software is remarkably successful especially in the light of the fact that no assumptions regarding the form of the size distribution are involved. Present size distributions obtained from the ultrasonic spectra appear to be broader than the corresponding Sedigraph results on the diluted samples.

Comments on Earlier Attempts to Use Ultrasound Attenuation Measurements of Slurries for Particle Sizing

The Armco Autometrics particle size monitor uses ultrasound attenuation measurements at two different frequencies to obtain a single point on the cumulative size distribution curve. Cushman⁹ et al. have described a similar apparatus. Both of these are limited to typically coarse (over 20 μm) and broad distributions encountered in mining ore grinding operations. Uusitalo¹⁰ et al combined measurements of attenuation and side scattering at a fixed angle. They claim improved accuracy but using their method they can determine the mean particle size only. Later ProAssist developed a technology, SDE-21¹¹, that is capable of providing five points on the cumulative distribution curve using ultrasound attenuation measurements at various frequencies over 1 to 6 MHz range. The theoretical expression relating the attenuation coefficient to frequency, particle size, and concentration used in the SDE-21 technology is of limited validity. Empirical calibration often is used to improve their accuracy. Their limitations arise from the use of a narrow operating frequency range, and an approximate, semi-empirical theoretical expression for the kernel function. These technologies, Armco Autometrics PSM and ProAssist SDE-

⁹ U. S. Patent No. 3,779,070 issued Dec. 18, 1973.

¹⁰ U. S. Patent No. 4,412,451 issued Nov. 1, 1983.

¹¹ Alba, Felix, private communication, July 1, 1987.

21, have been used successfully for mineral slurries containing particles with mean sizes ranging from 40 to 100 micrometers. Idea of using attenuation measurements at several frequencies to estimate particle size distributions had been described earlier by Riebel¹². Riebel envisioned using a range operating frequencies such that the wavelength corresponding to the largest frequency is approximately equal to the diameter of the smallest particle and the wavelength corresponding the lowest frequency is approximately equal to the diameter of the largest particle. He however did not address the underlying issues related to determination of $\alpha_{\text{mono}}(f, D)$, attenuation coefficient contribution at a frequency f due to a monodisperse population with all particles having diameter equal to D in suspensions of moderate to high particle concentrations. A few attempts of estimating PSD of concentrated emulsions using attenuation measurements over 100 kHz to 185 MHz have been reported. These were based on trial-and-error fit between the Allegra and Hawley theory and the experimental measurements. Significant progress was made recently by Alba¹³ in developing a technique for estimating particle size distributions of suspensions using ultrasonic spectroscopy. He combined Allegra and Hawley formulation with the Waterman and Truell multiple scattering results as suggested by Davis to obtain the necessary kernel functions. All of the above mentioned technologies do not adequately address the needs of slurries containing submicron particles at high volume fractions and can not address a broad variety of colloids encountered in industry.

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¹² U.S. Patent No. 4,706,509 issued Nov. 17, 1987.

¹³ U. S. Patent No. 5,121,629 issued June 16, 1992.