ENVIRONMENTAL AND WASTE MANAGEMENT: ADVANCEMENTS THROUGH THE ENVIRONMENTAL MANAGEMENT SCIENCE PROGRAM

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CHARACTERIZATION OF SOLID LIQUID SUSPENSIONS UTILIZING NON-INVASIVE ULTRASONIC MEASUREMENTS

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ABSTRACT

Rapid, on-line characterization of the particle size and concentration of moderate to highly concentrated slurries is required for the efficient and waste remediation at the DOE complexes. This paper discusses the advancements achieved under the Environmental Management Science Program to accurately characterizes high-level waste at the high concentrations expected at the DOE complexes. In addition, the results are applicable to efficient process measurement and control in many chemical and pharmaceutical manufacturing processes. Existing methods for determining the particle size and concentration of non-dilute slurries based on ultrasonic attenuation can become inaccurate due to the complex interactions of ultrasonic waves with the constituents of the slurries and the necessity for very careful transducer alignment. Two measurements that help to overcome these difficulties are the ultrasonic backscattering and diffuse field. The backscattering measurement is attractive because viscous, thermal and inertial effects have small contributions to the backscattering. In addition, the backscattering theories are simpler than attenuation theories and lend themselves to more stable inversion processes. Furthermore, the measurements of backscattering measurement does not require long travel distances and can be made with a single transducer thus eliminating alignment problems. We will present ultrasonic measurements and theoretical comparisons on solid liquid suspensions designed to elucidate the particle size and concentration at high concentration relevant to the high-level waste at the DOE complexes.

BACKGROUND

There are several properties of an ultrasonic field that elucidate the characteristics of the medium in which the wave is propagating. These properties include attenuation, backscattering, velocity and the characteristics of the diffuse field. These measurements have been applied for many years to study solid-liquid slurries and elastic solids with varying degrees of success. Performing the measurements of attenuation and velocity is a relatively straight forward task. The more difficult task is the absolute measurement of the backscattering and the theoretical development that accurately models the mechanisms contributing to the measured values. In addition, the inversion of the theories for comparison with experimental results to provide the specific characteristics of the slurry can be computationally intensive and inaccurate at high solids concentrations. When describing scattering in slurries, there are several important aspects, including, particle size and shape, concentration, inhomogeneities and the degree of multiple scattering present. In this brief review, we identify the gaps in technology that create obstacles to slurry characterization and describe how our proposed work will overcome those obstacles.

Attenuation has been used widely and successfully to characterize slurries at low levels of dilution. Allegra and Hawley [1972] provided the groundbreaking theoretical treatment for solid-liquid suspensions. Their model accounts for the attenuation due to viscous damping, as the particle moves and changes shape, the thermal loss as heat is exchanged between the ultrasonic field and the particle, and the scattering loss as the propagating wave is scattered at the interfaces between the fluid and the solid particles. They obtained good agreement between experimental measurements of attenuation and theoretical predictions only at low concentrations. Furthermore, their theory lacks an explicit term for the backscattering amplitude and does not incorporate particle size distributions and contributions from multiple scattering. These deficiencies hinder the applicability of devices that rely on this theory. The main obstacles for implementing attenuation measurements to slurry characterization have been proven to be the mathematical complexities of accounting for multiple scattering at high concentrations and the accompanying complex nature of the inversion process.

Backscattering measurements have several advantages over attenuation measurements, including insensitivity to alignment of the transducer and small propagation distances, thus making them ideal for characterizing highly attenuated slurries. In addition, since the direct backscattered field usually can be described by single-scattering processes, the mathematical inversion process is often more simple and stable than those used for attenuation.

Backscattering in slurries and suspensions has been less thoroughly studied, relative to attenuation, with the efforts focused on geologic and oceanographic applications. Notably, Hay and Mercer (1985) used the theory developed by Allegra and Hawley [1972] to explicitly determine the backscattering amplitude as a function of the elastic properties of the scatterers and the viscous fluid. They further extended their work to include particle size distributions of sand particles in suspension. In addition, He and Hay [1993] accounted for the irregular shapes of sand particles by performing an

ensemble average of the scattered pressure. They obtained good agreement with experimental results for sand particles in suspension up to k_a~1.

Another aspect of the backscattered field that is intimately related to the properties of the scattering media is the portion that undergoes multiple scattering and for which propagation can be described by a diffusion equation. While the majority of applications of the diffuse field have focused on characterizing solids, Weaver and Sachse [1995] focused their recent work on slurries. In their study, it was significant that measurements were performed on slurry containing strongly scattering glass spheres at a concentration of 62 wt%. In addition, Page *et al*. [1999, 2000] reproduced the work of Weaver and Sachse and then applied the diffuse field technique to measure particle motion in a non-stationary suspension. While these efforts show that the diffuse field can be used to characterize dense slurries, it is important to note that the theoretical predictions employed thus far have relied on a single scattering approach, creating poor agreement between theoretical and experimental results.

While these results are encouraging, past work has shown that the major obstacles for implementation are the inaccuracies in predictions and inversions at high concentrations. These findings indicate that single scattering models that predict attenuation and backscattering may be inadequate and that theoretical treatment of strong multiple scattering may be necessary, especially for k_a>1 and at high concentration. Also missing is the theoretical treatment of multiple size distributions. Many industrial processes contain multiple particle size distributions at high concentrations and overcoming these obstacles has been a major focus of our ongoing work.

EXPERIMENTAL METHODS

Velocity, Attenuation and Backscatter Measurements

Particle slurries with concentrations up to 40 weight percent (0, 5, 10, 15, 20, 30 and 40) were created by mixing glass particles (35 and 70 µm mean diameter) in de-ionized, degassed water. A cylindrical, 10.2-cm (4-in.) inside diameter, Teflon container containing opposing transducers as shown in Figure 1 was used to obtain velocity, attenuation, and backscatter measurements. A high speed mixer agitated the slurry to keep the particles in suspension during measurements. Velocity and attenuation measurements were obtained in a pitch-catch configuration with a pair of both 5-MHz and 10-MHz planar transducers. Deionized, degassed water was used to align the through-transmission transducers and also as a reference liquid for subsequent attenuation measurements. Backscatter measurements were obtained with four different transducers in pulse-echo mode: 5-MHz planar, 5-MHz focused at 0.79 cm (2.0 in.), 10-MHz planar and 10 MHz focused at 0.60 cm (1.5 in.) as shown in Figure 1.

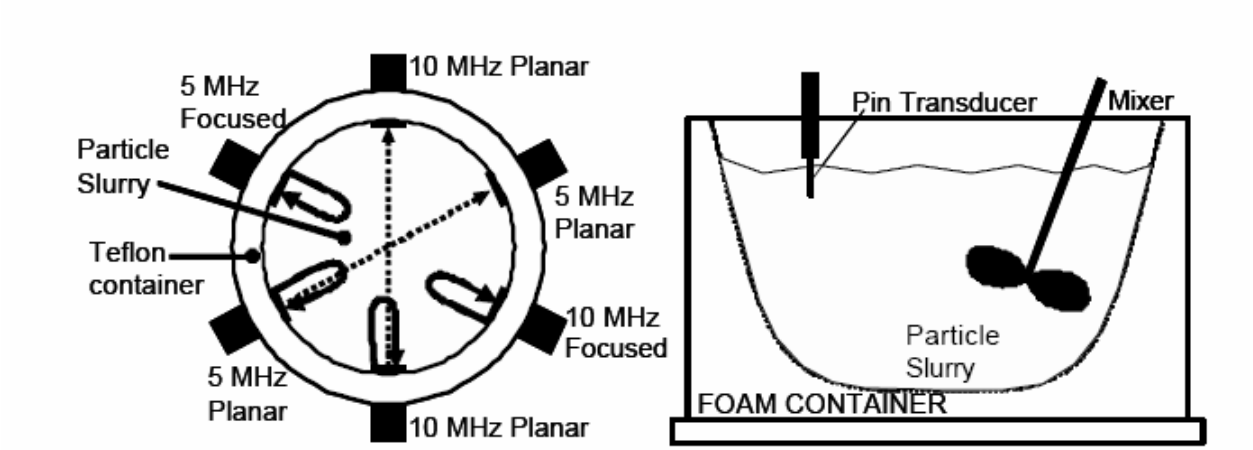

Figure 1. Top view of Teflon agitation chamber (left): particles were agitated to keep in suspension during testing; ring of transducers was used to propagate in both pitch-catch (dashed line) and pulse-echo (solid line) configurations. Diffuse field measurement setup (right) using a foam container and pin transducers.

A Ritec SP-801 pulser was used to excite the transducers and a RITEC BR-640 receiver was used to amplify and filter the received signals. The pulser was set to optimally excite the transducers with a square wave pulse dependent on the nominal transducer frequency response. The receiver gain was set to 64 dB for backscatter measurements and varied between -8 dB and 52 dB for through-transmission attenuation measurements. An input impedance of 50 Ohms was used with a bandpass filter between 1 and 12 MHz. For pulse-echo applications, a Ritec RDX-2 was used with a damping of 1300 Ohms and a low frequency cutoff of 1.6 MHz. Signals were captured at a sampling rate of 100 MHz, with a LeCroy 9310M oscilloscope and stored digitally on a computer *via* a GPIB communications port utilizing a Labview data acquisition program.

For pitch-catch velocity and attenuation measurements, 50 RF waveforms were averaged for each weight percentage. This signal was used to measure transit time (time to highest positive peak) and attenuation relative to water. The Fourier amplitude of each averaged signal was calculated, $\Gamma_S(f)$, and compared with a baseline Fourier amplitude from water, $\Gamma_{ref}(f)$, to calculate the attenuation as a function of frequency, $\alpha(f)$, using the following expression

$$\alpha(f) = \frac{1}{z}\ln\left[\frac{D_s(f)}{D_{ref}(f)}\frac{\Gamma_{ref}(f)}{\Gamma_s(f)}\right] \tag{1}$$

where z is the through transmission distance and D_s and D_{ref} are the beam diffraction corrections for sample and water reference, respectively. For these samples, the diffraction corrections were assumed to be the same for both the slurries and the water

reference. This assumption is reasonable considering that the velocity of the highest concentration slurry varies from water by only a few percent.

For backscattering measurements, 100 single-shot signals were captured at each concentration. The Fourier amplitude was calculated from a small time window of each RF waveform. The Fourier amplitude at each frequency was then averaged for all 100 waveforms. Since attenuation causes a diminution of the backscattered signals, this measure of the backscattering was corrected for the attenuation in the following manner. Each attenuation spectra was fit with a power law function within the frequency bandwidth of the transducer. This power law was then used to correct the backscattering signal.

Diffuse Field Measurements

A foam container was constructed to conduct diffuse field measurements (Figure 1). Particle weight percentages were the same as those used in the backscatter measurements. An ultrasonic setup consisting of a Ritec SP-801 pulser, BR-640 receiver were used to drive a Valpey-Fisher transducer, with a nominal diameter of 1 mm, in pulse-echo and pitch catch modes to measure the diffuse field signals. The pulser sent a 1-MHz, 400-volt square wave pulse to the transducer with a 25 Hz repetition rate. The diplexer was set at a damping of 1300 Ohms and a low frequency cutoff of 30 kHz. The receiver gain was set to 56 dB with a high input impedance and a bandpass filter between 500 kHz and 3 MHz. Five RF waveforms were averaged and captured at a sampling rate of 50 MHz. A joint time frequency analysis was performed on the received RF waveforms as detailed by Weaver and Sachse (1995). The decay rate of the diffuse field was determined as a function of frequency from the resultant amplitudes for each frequency window.

RESULTS AND CONCLUSIONS

Significant advancements have occurred in developing the experimental measurements of backscattering and diffuse fields. These novel measurements will enable for the characterization of solid liquid suspensions at higher concentrations than commercially available systems. Figure 2 shows the attenuation as a function of frequency for 35 µm and 70 µm glass spheres at concentrations up to 40 wt%. The attenuation as a function of concentration is shown in Figure 3, where the higher the concentration and particle size, the higher the attenuation. In Figure 4 the attenuation corrected backscattering is seen to increase as a function of concentration for both the 35 µm and the 70 µm glass spheres. Diffuse field results are shown in Figure 5, where the “energy” as a function of time is seen to decay with increasing decay rates as the concentration increased. The decay rate as a function of concentration is shown in Figure 5b. The results for these ultrasonic measurements are consistent with theoretical predictions as is shown in Figure 6 for the attenuation and backscattering.

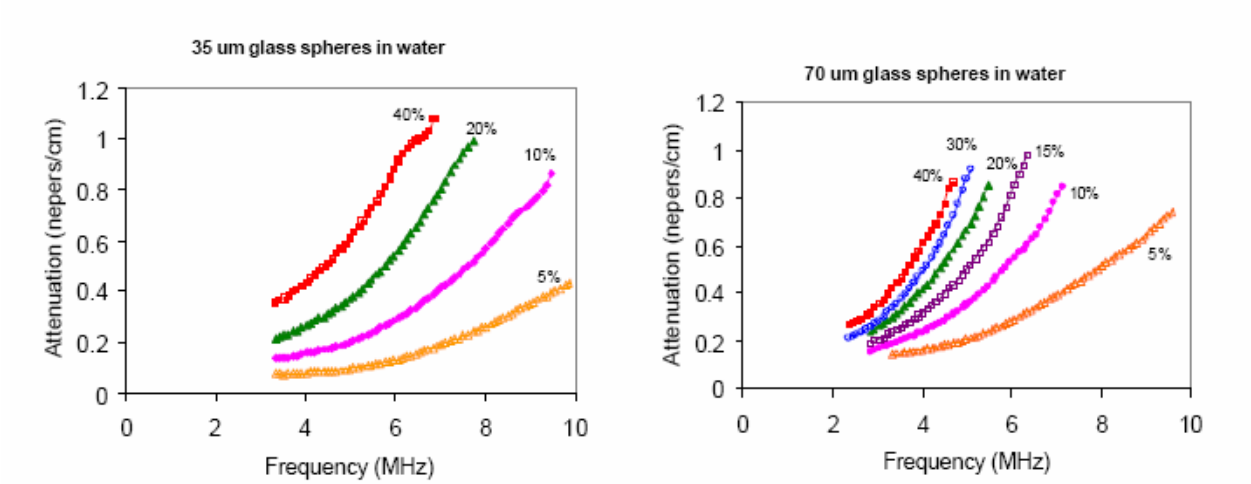

Figure 2. Attenuation as a function of frequency for 35 µm diameter glass spheres (left) and 70 µm glass spheres (right) for concentrations up to 40 wt%.

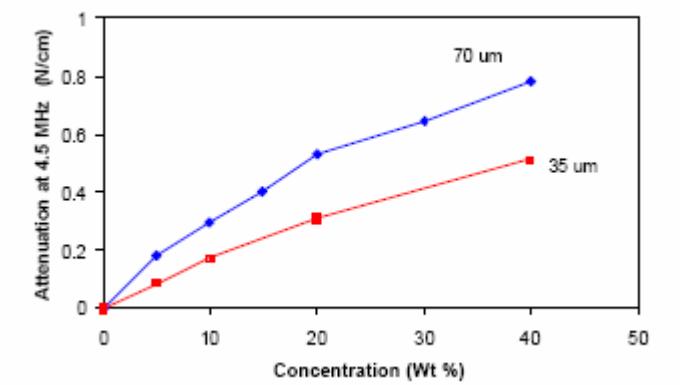

Figure 3. Attenuation at 4.5 MHz as a function of concentration for the 70 µm and 35 µm glass spheres.

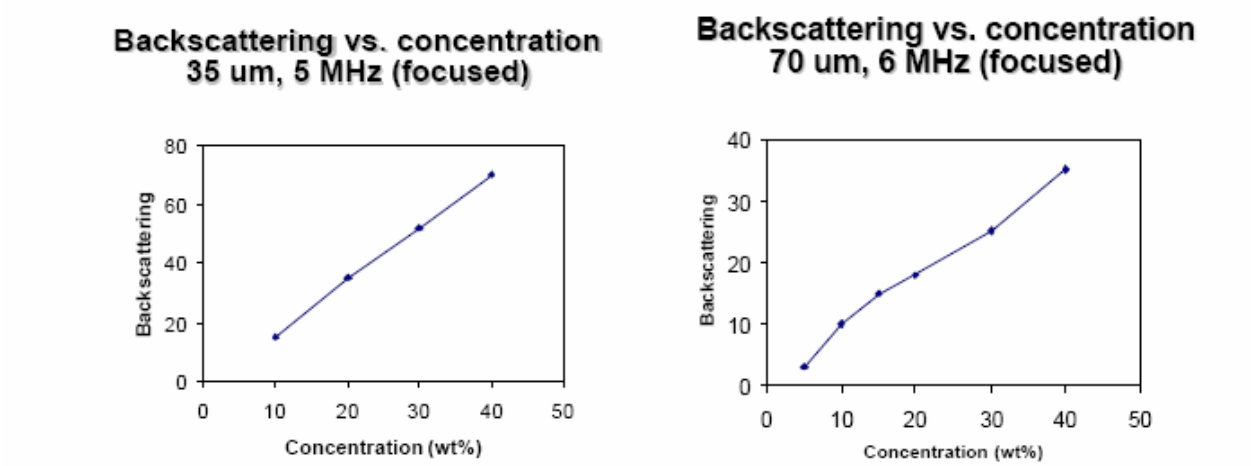

Figure 4. An experimental measure of the backscattering as a function of concentration for 35 µm diameter glass spheres (left) and 70 µm glass spheres (right) at concentrations up to 40 wt%

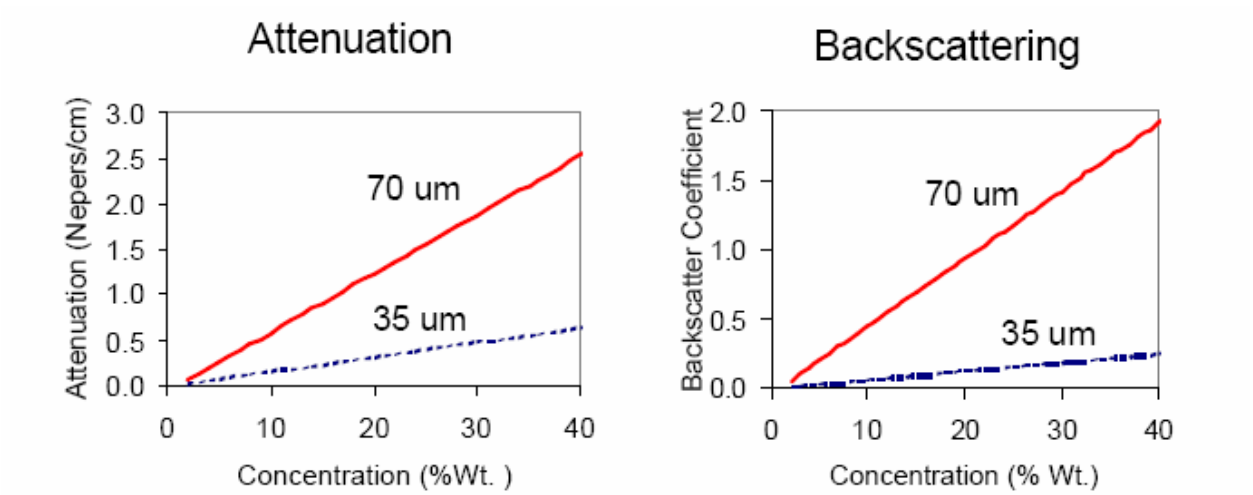

Figure 5. Theoretical predictions of attenuation (left) and backscattering coefficient (right) as a function of concentration for glass spheres for concentrations up to 40 wt%.

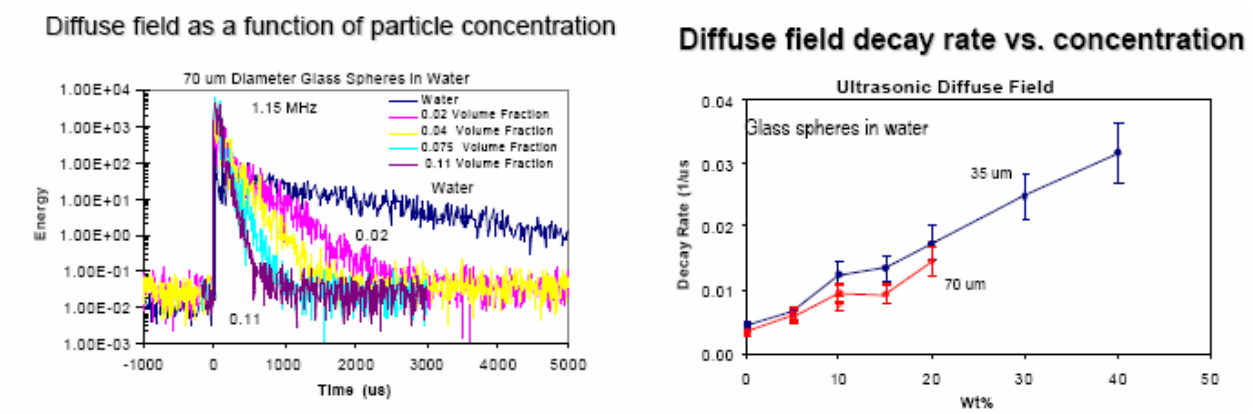

Figure 6. Diffuse field decay in particle slurries of varying concentration, from water to 0.11 volume fraction (left) and diffuse field decay rate as a function of concentration for 35 and 70 µm particles (right)

ACKNOWLEDGEMENTS

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REFERENCES

Allegra, J.R. and S.A. Hawley. 1972. "Attenuation of Sound in Suspensions and Emulsions: Theory and Experiments." *The Journal of the Acoustical Society of America*, **51**(5):1545-1564.

Hay, A.E. and D.G. Mercer. 1985. "On the Theory of Sound Scattering and Viscous Absorption in Aqueous Suspensions at Medium and Short Wavelengths." *Journal of the Acoustical Society of America*, **78**(5):1761.

He, D. and A.E. Hay. 1993. “Broadband Measurements of the Acoustic Backscatter Cross Section of Sand Particles in Suspension.” *Journal of the Acoustical Society of America*, **94**(4):2247.

Page, J.H., I.P. Jones, H.P. Schriemer, M.L. Cowan, P. Sheng and D.A. Weitz. 1999. “Diffusive Transport of Acoustic Waves in Strongly Scattering Media.” *Physica B*, **263-264**:37-39.

Page, J.H., M.L. Cowan, P. Sheng and D.A. Weitz. 2000. “Diffusing Acoustic Wave Spectroscopy of Fluidized Suspensions.” *Physica B*, **279**:130-133.

Weaver, R. and W. Sachse. 1995. “Diffusion of Ultrasound in a Glass Bead Slurry.” *Journal of the Acoustical Society of America*, **97**:2094-2102.