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Investigating Ultrasonic Diffraction Grating Spectroscopy and Reflection Techniques for Characterizing Slurry Properties

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Research Objectives

The particle size of a slurry and the viscosity of a liquid or slurry are both difficult to measure on-line and in real time. The objectives of this research are to develop the following methods for such measurements: 1) ultrasonic diffraction grating spectroscopy (UDGS) to measure the particle size of a slurry, 2) UDGS to measure the velocity of sound in a slurry using reflection from a grating as opposed to ultrasound traveling through a possibly dense slurry, 3) develop theoretical models and computer codes to describe the passage of ultrasound through a grating surface in order to increase the sensitivity of the particle size measurement, and 4) shear wave reflection techniques to measure the viscosity of a slurry.

Research Progress and Implications

This report summarizes research in the first year of a two-year EMSP renewal project, (following the three-year EMSP project that was completed on September 30, 2004). As described in last year's Annual Report, the effect of particle size in experiments with UDGS is the *first observation* of such an effect using ultrasonics. During this year experiments were performed using an aluminum grating with slurries of polystyrene spheres in water at a frequency of 3.5 MHz. The results show that slurries of 215 microns, 275 microns, 363 microns, and 463 microns have widely separated amplitudes, thus leading to identification of particle size. An important finding this year was that the experimental data compare very well with calculations based upon the inertial model for sound absorption in slurries. Calculations were also carried out to investigate the penetration of ultrasound into the slurry. These results are described below. Viscosity measurements for sugar water solutions were obtained with a fused silica wedge, having inclined sides at 45°. These measurements were made on-line and in real time using a data acquisition

system, in which the Matlab coding automatically changed the pulser and receiver settings. The viscosity values were in good agreement with those obtained independently using a laboratory viscometer.

Experimental measurements for UDGS

A schematic diagram of the data acquisition system is shown in Fig. 1. Ultrasound from the send transducer travels through the aluminum and strikes the grating-slurry interface, where it is diffracted into a number of reflected and transmitted waves, each characterized by an integer spectral order m. The specularly reflected wave (m=0) is received by the receiving transducer. The frequency of the toneburst signal (10 cycle sine wave) is incremented in 0.05 MHz steps over a frequency range. The maximum amplitude of the received signal is recorded for each frequency. The data consist of this amplitude plotted versus the frequency. As the frequency decreases, the angle of m = 1 longitudinal wave in the slurry increases and eventually reaches 90° and becomes evanescent, at the so-called critical frequency. Below this critical frequency, this wave cannot exist in the slurry and its energy is shared by all other types of waves. Hence, this is why we see a peak in the wave reflected by the grating. The evanescent wave consists of a wave traveling parallel to the grating surface, but its amplitude decreases exponentially with distance from the grating surface. It is somewhat akin to a surface wave. This evanescent wave interacts with particles in the slurry and energy is lost. This energy loss is dependent upon the particle size.

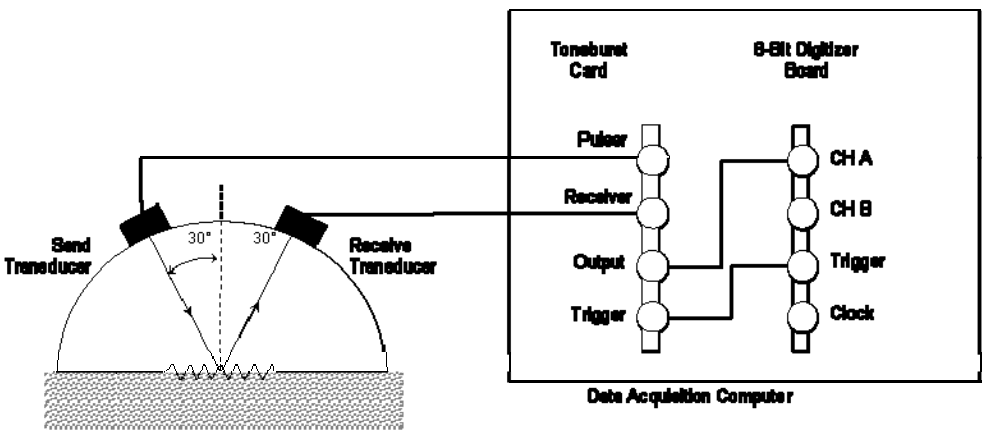

Fig. 1. Schematic diagram of the experimental apparatus for ultrasonic diffraction grating spectroscopy.

The data in Fig. 2 and Fig. 3 show the comparison of various concentrations of 215 micron polystyrene spheres in water at the critical frequency. Similar data are shown in Fig. 4 for 463 micron polystyrene spheres in water. Four slurries at a concentration of 11 Wt% are compared with water in Fig. 5. Here we clearly see the different behavior for the slurries of different particle size, showing the ability of UDGS to distinguish particle size.

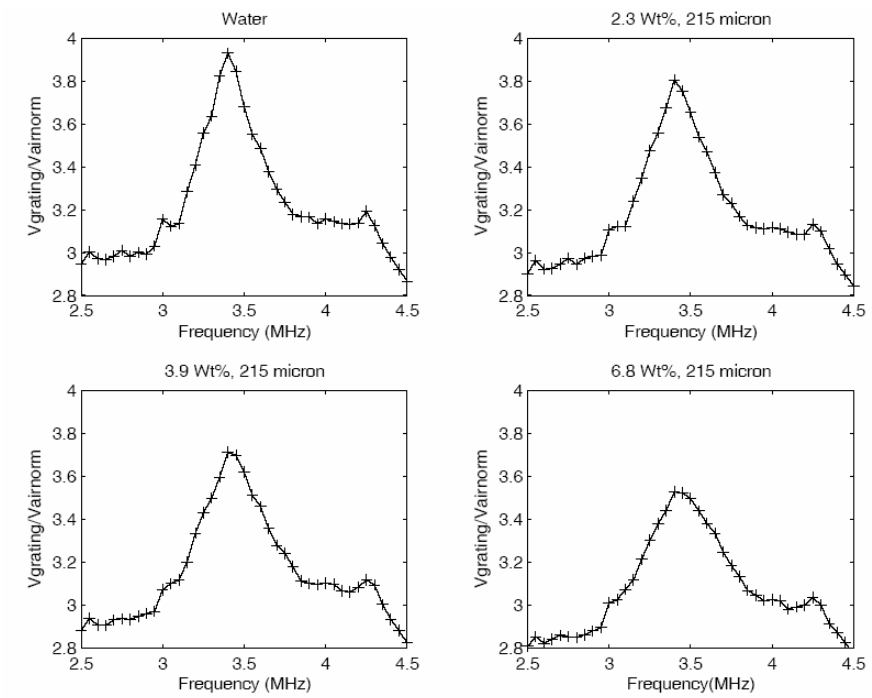

Fig. 2. Data obtained by the receive transducer for water and 215-micron diameter polystyrene spheres in water for weight percentages of 2.3 Wt%, 3.9 Wt%, and 6.8 Wt%.

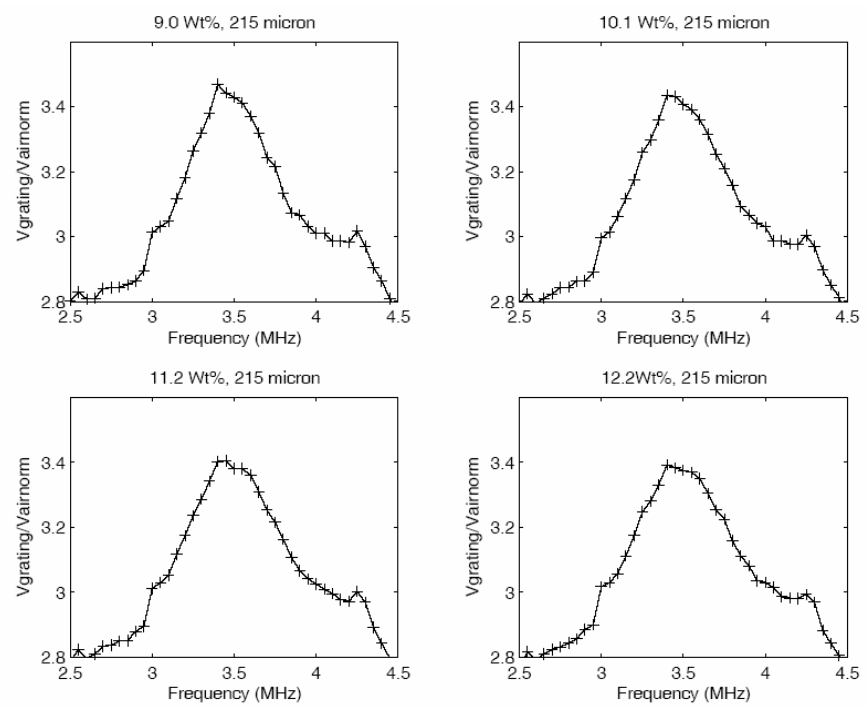

Fig. 3. Data obtained by the receive transducer for water and 215-micron diameter polystyrene spheres in water for weight percentages 9.0 Wt%, 10.1 Wt%, 11.2 Wt%, and 12.2 WT%.

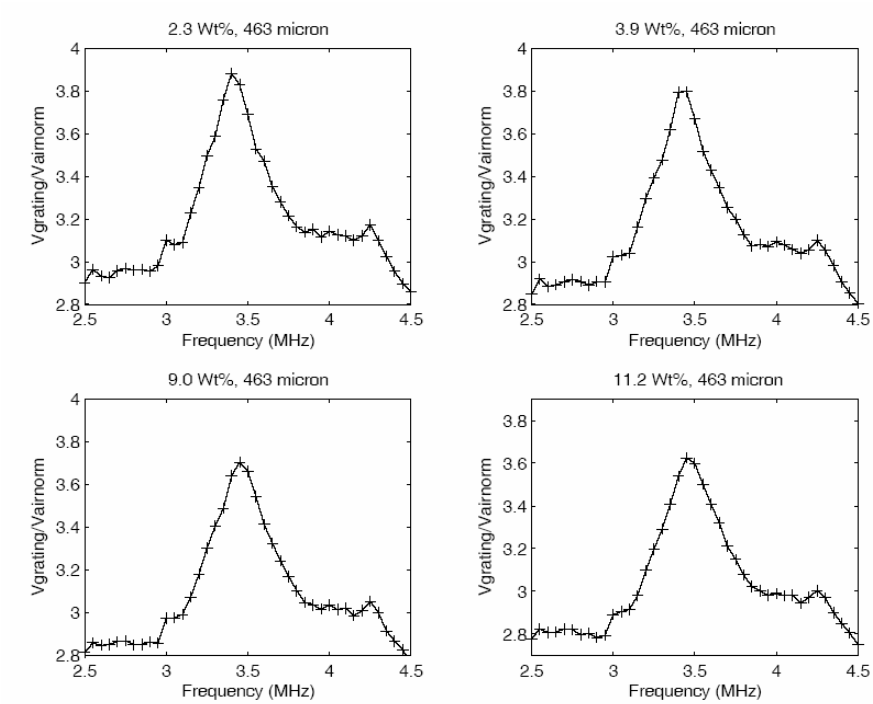

Fig. 4. Data obtained by the receive transducer for 463-micron diameter polystyrene spheres in water for weight percentages of 2.3 Wt%, 3.9 Wt%, 9.0 Wt%, and 11.2% Wt%.

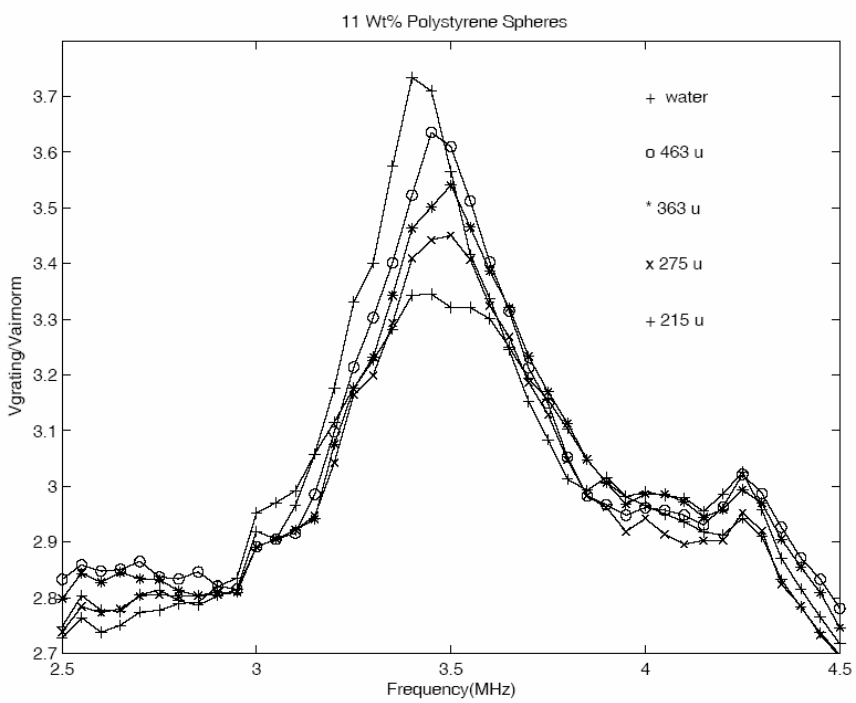

Fig. 5. Comparison of data of water and slurries of polystyrene spheres at 11 Wt% for sphere diameters of 215 microns, 275 microns 363 microns, and 463 microns.

In Fig. 6 and Fig. 7 the data are normalized by dividing the peak height for the slurry at the critical frequency by that for water and plotted versus the volume fraction of the slurry. Theoretical calculations were carried out using the inertial model of Atkinson and Kytomaa [1] for sound absorption and are shown as solid or dashed lines. Since the particles were obtained by using sieves of various sizes, the calculations were carried out for the lower and upper limits of the particle size for a given slurry.

An important consideration is the penetration of the ultrasound into the slurry. The toneburst signal has a small bandwidth, but still contains a small range of frequencies. This range of frequencies was considered and the results for the amplitude of the evanescent wave as a function of the perpendicular distance z from the grating surface are shown in Fig. 8. This shows that the slurry penetrates to a distance larger than 5 mm.

The objective of the theoretical calculations is to shift a large amount of the available energy to the m = 1 transmitted longitudinal wave in the slurry, which will produce a larger reflected signal. Experiments can then be carried out using the apparatus designed from the predictions of theoretical calculations.

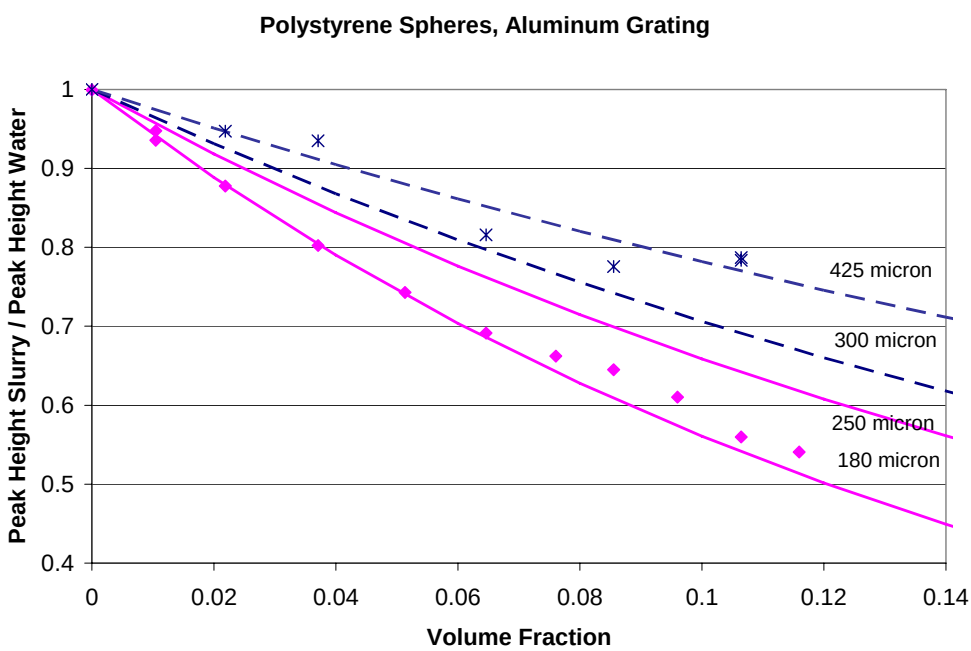

Fig. 6 Data obtained for the ratio of the peak height for the slurry to that for water for slurries of 215-micron diameter spheres and 363-micron diameter spheres. The solid and dashed lines show the comparison of the data with the inertial model for the upper and lower limits of the particle size distributions.

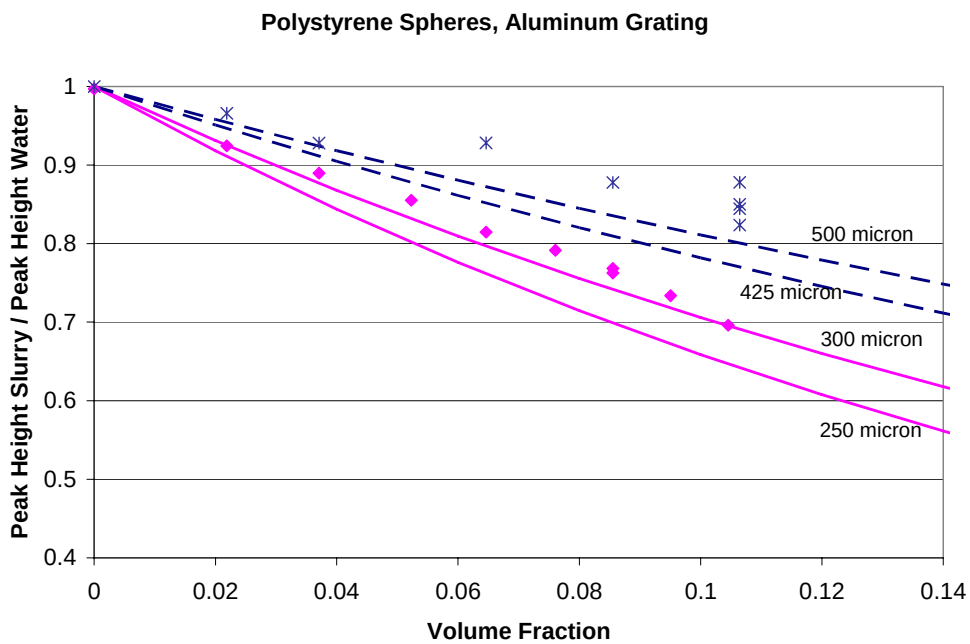

Fig. 7. Data obtained for the ratio of the peak height for the slurry to that for water for slurries of 275-micron diameter spheres and 463-micron diameter spheres. The solid and dashed lines show the comparison of the data with the inertial model for the upper and lower limits of the particle size distributions.

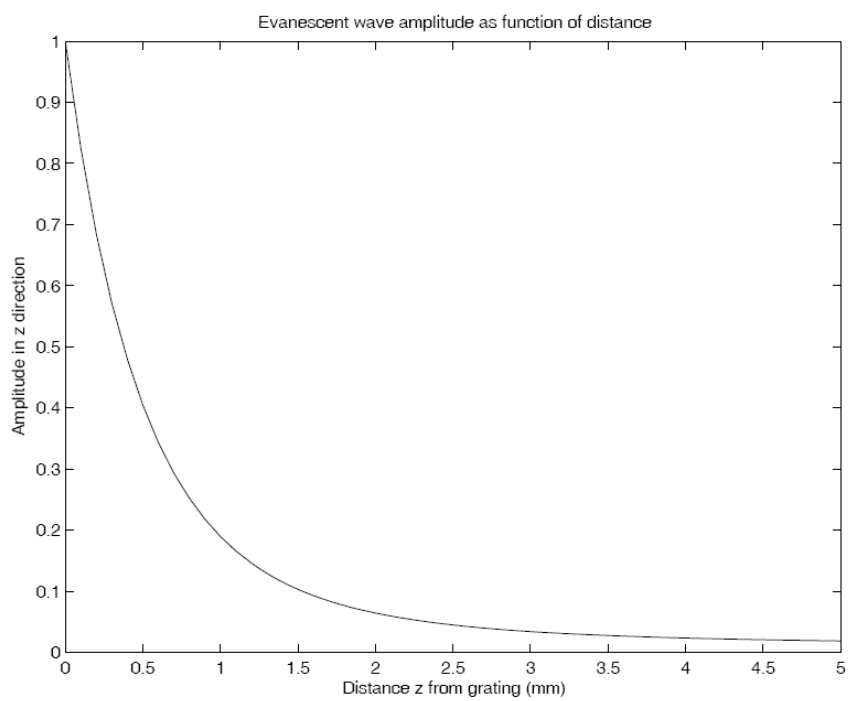

Fig. 8. Effective amplitude of evanescent wave versus perpendicular distance from grating surface.

Theoretical and computer modeling

The passage of ultrasound through a grating surface between a solid and a fluid medium is investigated theoretically using the mathematical formulation of Mampaert and Leroy [2]. These studies were for perpendicular incidence and are being extended to include any angle of incidence. The effects of different shapes of the grooves of the diffraction grating and the effects of the angle of incidence of the beam as it strikes the interface can be easily studied by this method. This formulation can be extended relatively easily to accommodate the finite width of the incident ultrasonic beam.

In this formulation, the acoustic field is described in terms of the velocity potential. The diffracted field is described by an infinite sum of plane waves. In the calculation the sum is limited to M, which is suitably large. This description is valid 1) when the peak-to-valley height H of the grating << the wavelength in the solid medium λi and also 2) when the grating spacing d is comparable to λi . Numerical instabilities occur if any of these restrictions are violated. Specifically, if d is not comparable to λi or $H/\lambda i > 0.15$, the solution does not converge with sufficiently large M. In our calculations, we have observed that M = 20 is sufficiently large to represent converged solutions when the limits of the validity of the mathematical theory are not violated.

We have calculated the reflection and transmission coefficient when an incident longitudinal wave strikes the saw-toothed interface between aluminum and water at varying angles of incidence. We have chosen a grating spacing d of 500 microns. At an incident angle of 45°, the critical frequency is 3.55 MHz and at 0°, it is 2.94 MHz. The wavelength λi in aluminum at this frequency is 1780 microns. The height H of the grating is chosen to be 50 microns. These values satisfy the restrictions described above.

In the experimental set up in Fig. 1, the reflection of the incident longitudinal wave to the receive transducer corresponds to reflection of order m = 0. Fig. 9 shows the calculation of this reflection coefficient |D0| as a function of frequency for 10 angles of incidence. Note that the critical frequencies at incident angles of 0° and 45° are at 2.94 MHz and 3.55 MHz, as expected. It is important to note the sharp increase in the reflection coefficient at incident angles of 39° and 45°. This is the question that we need to answer: Under what conditions do we find the largest reflected signal?

The peak in the reflected signal is due to energy conservation as the m = 1 transmitted longitudinal beam in the liquid or slurry reaches 90°, becomes evanescent, and then ceases to exist in the liquid. Some of the energy is transferred to the reflected signal. Therefore, where the reflected signal peaks, we also expect a peak in the m = 1 transmitted signal. Fig. 10 shows this to be the case. The reflection coefficient |D0| in Fig. 9 and the transmission coefficient |T1| in Fig. 10 are much larger at 39° and 45° than at other incident angles. Such information is essential in designing an efficient apparatus for the measurement of particle size.

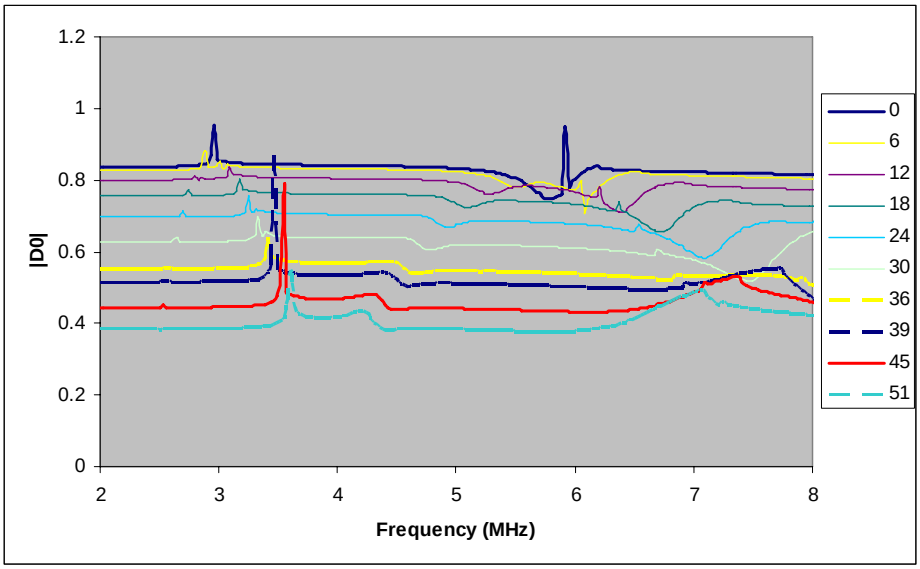

Fig. 9. Computer modeling for the reflection coefficient versus frequency, which shows the effects of the angle of incidence for the beam striking the grating surface.

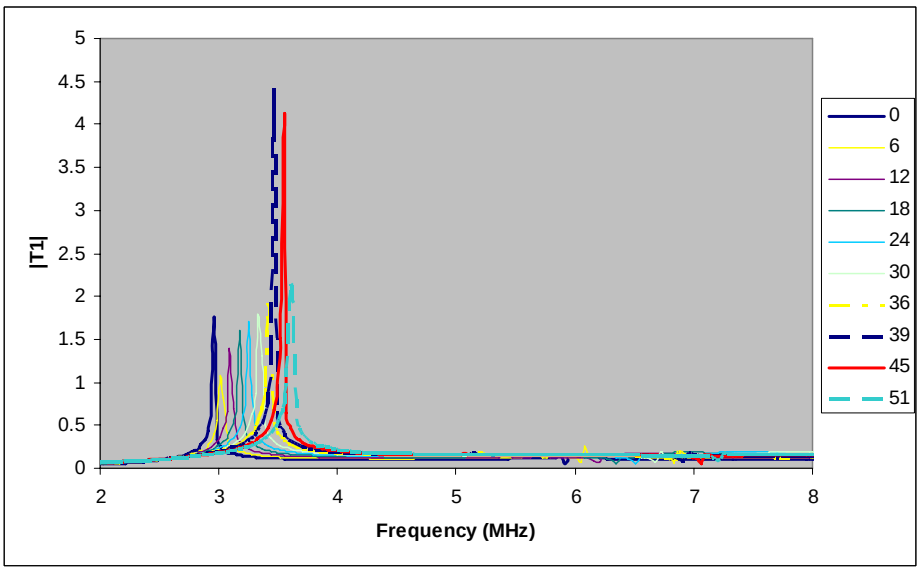

Fig. 10. Computer modeling for the transmission coefficient versus frequency, which shows the effect of the angle of incidence for the beam striking the grating surface.

Viscosity

The measurement of viscosity uses the reflection of a shear wave at the solid-liquid (slurry) interface. The theory is well-known, but interestingly, has not been very well developed experimentally. Previously, only special applications were considered. We developed an on-line real time measurement using a computer-controlled data acquisition system. The coding was written using the Matlab language. Fig. 11 shows the experimental setup for measuring viscosity for Newtonian fluids and the shear modulus for non-Newtonian fluids. Either parameter can be used to detect changes in the liquid/slurry for process control. The transducer produces a shear wave that travels to the quartz-liquid interface and then reflects

from the other side of the wedge. The ultrasound makes more than 20 round-trips between the two inclined sides of the wedge and the interface. Each time the ultrasound strikes the interface, a small amount of ultrasound travels into the liquid and these echoes amplify the effect. The ability of liquid to support ultrasonic shear waves increases with the liquid's viscosity. Thus, by measuring the reflection coefficient at the interface, the viscosity can be determined. Fig. 12 shows the comparison of the viscosity of sugar water (SW) solutions using the ultrasonic sensor with those obtained by independent laboratory methods. The reflection coefficient range in value from 0.975 to 0.996 for the SW solutions shown in Fig. 12. Very high accuracy is required for such a measurement. Such a measurement is possible due to the self-calibrating method developed at PNNL, for which U. S. patent 6,763,698 was awarded in July, 2004. This means that, if the pulser voltage changes, the measurement is not affected because each echo is affected in the same way. An analysis method was developed which capitalizes on this effect and results in an accurate measurement of the reflection coefficient, and hence, viscosity.

These measurements were obtained using a wedge having 45° angles, in order to reduce the size of the wedge. Previous measurements were obtained using a wedge having 70° angles. The sensitivity is larger for the 70° wedge, but it is twice as long. The same data acquisition system was used for both sensors.

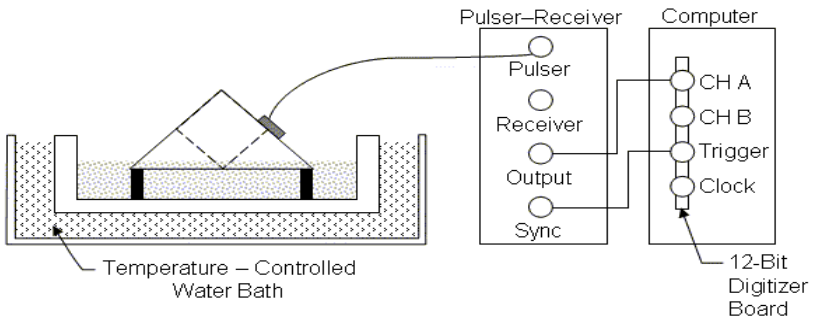

Fig. 11. Schematic diagram of the experimental apparatus for the measurement of viscosity on-line and in real time.

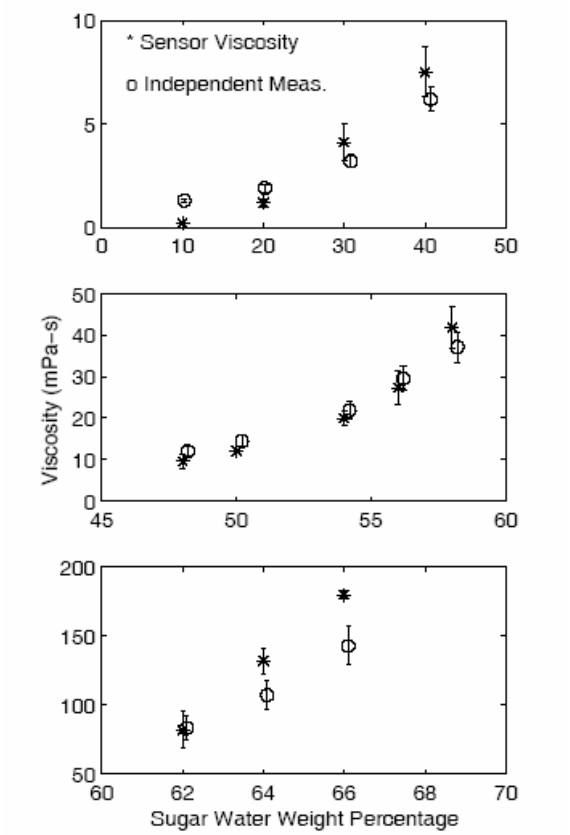

Fig. 12. Comparison of viscosity for the ultrasonic sensor with that obtained independently with a laboratory viscometer.

Implications

The important implications of our EMSP research is that, when fully developed, UDGS will enable the measurement of the slurry particle size on-line and in real time. The experimental method is straight-forward and so is the analysis. The grating surface can form part of the pipeline wall or be inserted into a tank. Further, such an instrument would be quite small. The measurement of viscosity using a fused quartz wedge agree with independent laboratory measurements. We have a system that can measure the viscosity-density product on-line and in real time *now*. Coupled with a measurement of density, such as the density sensor developed at PNNL, the viscosity can be determined. The base of the quartz wedge can form part of the pipeline wall. A system can be developed so that the electronics are smaller (perhaps a single board for the computer, rather than separate pulser and digitizer boards) and user-friendly. These techniques for making a surface part of the pipeline wall have been developed for the density sensor installed on the pipeline between SY101 and SY102 at the Hanford Site, in which the base of a plastic wedge formed part of the pipeline wall.

Planned Activities

Currently, an aluminum grating apparatus is being designed, using a frequency of 7.5 MHz, to insonify a large area of the grating surface. The resulting peak width is expected to be narrower and thus, increase the sensitivity of the particle size measurement. The incident angle of the ultrasonic beam upon the grating surface will be chosen based upon the theoretical modeling calculations. These experiments will takes place during the next 3 months. Based upon these results, the next series of experiments will be planned. The objective is to increase the range of the particle size measurement from 5 microns to 200 microns.

The modeling, extended from that of Mampaert and Leroy, brings out the essential features of plane wave interactions with a periodic interface. During the next year, Dr. Ahmed will incorporate into it 1) the effect of the finite beam width and 2) the effect of viscosity of the fluids. As described earlier, there is a limitation to the validity of the computer model. Therefore, he has begun to develop a finite-volume numerical scheme which will allow us to investigate the total displacement fields before and after interactions with a non-planar interface between two media. As a first step toward that goal, he has completed the numerical scheme for a planar interface between a solid and fluid.

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