Project 94676

Physical Characterization of Solid-Liquid Slurries at High Weight Fractions Utilizing Optical and Ultrasonic Methods

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RESULTS TO DATE: Research Objective The goal of this proposed work is to directly address the need for rapid on-line characterization of the physical properties of HLW slurries during all phases of the remediation process, from in-tank characterization of sediments to monitoring of the concentration, particle size, and degree of agglomeration and gelation of slurries during transport. There are three tasks: 1) develop new optical and acoustic scattering measurements to provide the fundamental science needed for successful device development and implementation, 2) develop theories that describe the interrelationship between wave propagation and the physical properties of the slurry, and 3) perform inversions of the theories and compare them with the experimental measurements to non-intrusively characterize slurries.

Research Progress and Implications

Our studies of high concentration slurries (up to 40 wt %) utilizing both optical and ultrasonic fields are ongoing. Slurries with mean particle sizes from ~0.01um to 800 um have been studied at concentration ranging from very dilute (< 1 wt %) to 40 wt %. The interrelationships between the attenuation, the direct backscattered field, and the diffusely propagating field have been investigated. This integration of both optic and ultrasonic techniques is unique and exciting because it offers a combined methodology that exploits the best features of these two powerful techniques. The teaming of significant expertise in both experimental and theoretical optics (University of Washington) and in experimental and theoretical ultrasonics (PNNL) has provided a synergistic approach to coupling these complementary techniques. This combination of capabilities has allowed characterization to be performed over a broader range of particle sizes and concentrations than can be accomplished with existing techniques.

Optical Measurements

The optical measurements are based on the investigation of slurries with Optical Low-Coherence Reflectometry (OLCR). OLCR is a white-light interferometric technique that incorporates a broadband light source with a classical Michelson interferometer. Initial work was performed using a Hewlett-Packard OLCR instrument and evolved to use a more flexible instrument developed by Optiphase. We have designed a probe for solid-liquid slurries in which the amplitude ratio of the reflection from multiple interfaces can be related to the refractive index of the sample. In the Rayleigh scattering regime, the refractive index can be directly related to the concentration of dispersed material of known refractive index, and can be diagnostic during agglomeration as particles transition from one scattering regime to the other. This feature aids us in deconvoluting dynamic changes in the slurry system.

Advances in the theory of coherent multiple scattering have been made in optics and acoustics, specifically to begin to extract information pertaining to system properties, such as particle size, concentration, and polydispersity. A manuscript is in preparation, in which, the development of theoretical expressions in optics based on the introduction of an additional coordinate and functional integral averaging is compared with experimental data and the inverse problem of characterization of multiscattering media by coherent backscattering measurements is considered.

One of the most interesting results of this work is the observation of so-called Mie resonances when the particle radii of a randomly distributed system are comparable with the wavelength of light. This non-monotonic oscillation of backscattered intensity is dependent upon optical parameters, such as effective dielectric contrast between the medium and the particles. A manuscript has been published to describe both the experimental and theoretical Mie resonance results.

Both standard polystyrene spheres and DOE waste simulants have been used for polydispersed measurements. We have reported on OLCR measurement capabilities in a more complex and realistic matrices including bimodal mixtures of individual polydispersed alumina suspensions. These were easily distinguishable from each other when measured individually at 30 wt %, and the trend of changing average particle size can be easily followed through the profile amplitude. To support our theoretical work, we have used as a model the analysis of bimodal monodispersed polystyrene mixtures. The decay curves of the resulting OLCR profiles are very similar in intensity and slope and clear relationships between the contributions from the bimodal components and the regions of the profiles cannot be easily determined. Differences between bimodal system compositions arise however, when the fluctuations of an individual signal from the mean are analyzed throughout the entire OLCR profile. These fluctuations are not noise-based and do not average out with multiple scans in the same location of a solid sample. Rather, they are due to fixed heterogeneities within the system and contain information regarding system distribution and dynamics. Variation from the mean signal is measured over each scan over the decay length of the profile. Then, by "stepping through" the OLCR profiles and correlating the intensity of individual data points to their neighbors, a sense of a correlation distance is found which is related to the composition of the matrices.

Fluctuations are especially important for monitoring phase transitions, such as gelation. The OLCR profile of a liquid solution is relatively smooth, due to Brownian motion causing an averaging of the signal fluctuations. However, as a sample transitions into a more solid state, Brownian motion slows and the fluctuations in the OLCR profile increase. However, as a sample transitions into a more solid state, Brownian motion slows and the fluctuations in the OLCR profile increase. This is because only limited numbers of random particle distributions are available for analysis by the wavepacket. When the system under test becomes solid, only one distribution is available and the fluctuations will become very stable. Gelation, agglomeration, and sedimentation have all been shown to occur by adjusting parameters of the alumina system (i.e. pH, flow rate, temperature) and are being systematically studied.

Overall, we have made significant progress in the analysis of optical signals obtained from multiple scattering events. We have observed the appearance of Mie resonances, a non-monotonic oscillation of backscattered intensity dependent on optical parameters, over the interval where radii are comparable to the wavelength. The results above clearly indicate that it is possible to extract important static and dynamic parametric information from dense scattering systems with OLCR. Between the refractive index information, fluctuation distributions, amplitude variation, and decay rate, we should be able to extract characteristic information about particle systems. If all the variables are changing simultaneous, we hope to further resolve the parameter convolution by utilizing the synergy of the project and incorporating information from the ultrasound data.

1. A. M. Brodsky, S. L. Randall, and L. W. Burgess "Coherence Effects In Light Backscattering By Multiscattering Media", (Manuscript for Physical Review in preparation)

2. S. L. Randall, A. M. Brodsky, and L. W. Burgess "Manifestation of Mie Resonances in Light Scattering from Multiscattering Media", Inter. J. of Mod. Phys B, V. 19, No. 4 (2005)