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Polydisperse particle size characterization by ultrasonic attenuation spectroscopy in the micrometer range

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Abstract

The theoretical advantages of ultrasonic attenuation spectroscopy for particle size are currently not fully utilized. Especially in the region of larger particles, there is a lack of experimental confirmation of applicable models which may be used to infer particle sizes from measured attenuation spectra. With the present work, an attempt is made to supply experimental data, obtained with a commercially available ultrasonic attenuation spectrometer, and model calculations, which are based on the resonant scattering theory. It is shown that measured attenuation results for various combinations of disperse and continuous phase for both polydisperse emulsions and suspensions are reproducible by calculation.

The approach is further examined for suspensions of porous particles. Here, the resonant scattering approach is combined with the Biot model for poroelasticity to obtain attenuation results with several fractions of titania aggregates, differing in particle size and pore diameter.

The results indicate that the theory of resonant scattering is a valid approach if applied to particle size characterization in the large particle limit.

Keywords: Attenuation spectroscopy; Particle size characterization; Resonant scattering theory; Porous particles

1. Introduction

For many of the steps in microelectronics manufacturing or for process or environmental control, the size and the concentration of a dispersed particulate phase is of importance. Whether it is for determining the minimum or maximum size of particles present or the distribution of particle sizes in a process, adequate measurement techniques should be employed. The use of ultrasound for this purposes has gained attention over recent years. One measurement technology based on the evaluation of sound signals is the ultrasonic attenuation spectroscopy. It is based on the known relation between the size and concentration of particles in a dispersion and the attenuation of a sound wave of known frequency passed through the dispersion. Its applicability has been demonstrated for processes such as milling, slurry characterization, and ceramics and food processing. Ultrasonic attenuation spectroscopy has advantages over other methods because it allows for measurements under production conditions due to the fact that signals for the analysis can be obtained even for highly concentrated dispersion.

While commercially available ultrasonic attenuation spectrometers have been obtainable for a number of years, their potential has not been fully used. One problematic field of application is the size region for particles or droplets larger than 10 μm. Another area of concern exists for aspherically shaped or inhomogeneous particles. The reason for the former problem is that there are no established and confirmed models for the size region of larger particles. With respect to systems of inhomogeneous particles the difficulties with the method stem from the ''spherical and homogeneous particle'' assumption of all models used for analyzing the size parameters.

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This work tries to address the shortcomings of ultrasonic attenuation spectroscopy theory for larger particles, emulsion droplets and inhomogeneous particles by comparing measurements obtained with a commercially available ultrasonic attenuation spectrometer to model calculations. The predictions were obtained from models which have been applied in other fields of ultrasonics research for a long time but have not been confirmed for particle characterization applications. With the incorporation of these results in analysis software, an extension of the measurement range of ultrasonic attenuation spectroscopy could be possible.

All measurements were carried out for disperse systems of diverse acoustic contrast to confirm the applicability. Size distributions and material properties were obtained by methods independently of ultrasonic attenuation spectroscopy in order to confirm the validity of the approach.

2. Ultrasonic attenuation spectroscopy for particle size analysis

2.1. Measurement setup

Attenuation spectroscopy is based on analyzing the amplitude of a sound wave passed through a measurement chamber containing a dispersion [1,2]. The sound wave originates from one transducer, and for the case of a transmission setup, is detected by a second transducer. From the wave amplitude at the beginning ($\hat{p}_0$) and after passing through the measurement chamber ($\hat{p}$) of length l, the attenuation coefficient α can be calculated [3–5]:

$$\alpha = -\frac{1}{l}\ln\frac{\hat{p}}{\hat{p}_0} \tag{1}$$

For this study, an attenuation spectrometer DT-1200 (dispersion technology) was used. The device contains a measurement chamber with a pair of broadband transducers. One of the transducers is fixed; the other (movable) transducer is used to set different gap widths l. This setup makes use of the fact that different attenuation levels can be measured for different materials. For some materials, a large gap width may be necessary to detect a measurable wave attenuation by the dispersion, and for others a smaller width is required because attenuation is too high. Additionally, an influence of frequency can be observed, since higher wave frequencies are absorbed much more easily. The device is able to cycle through a series of user-specifiable discrete frequencies in the range from 3 to 99 MHz. A time-of-flight analysis for sound velocity measurements at one frequency is performed in addition to the evaluation of signal strengths $\hat{p}$ and $\hat{p}_0$. The result that is made available for further evaluation is a vector of the attenuation values α at each of the selected frequencies, and a speed of sound measurement. For the present study, the data were used as supplied by the device, that is, without alteration of the signal processing steps.

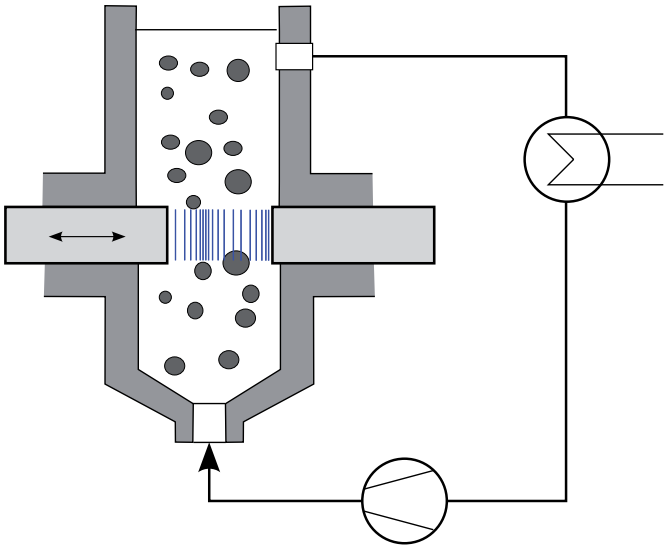

Fig. 1. Attenuation measurement setup base for suspensions based on a DT 1200 attenuation spectrometer with heat exchanger and suspension pump.

The standard measurement chamber of the DT 1200 contains a magnetic stirrer for homogenizing. This is sufficient for suspensions of nano-scaled particles, but for larger particles, the higher settling velocities often outweigh the homogenization. Thus, the suspension flow was intensified by installing a tubing pump which conveys the suspension upwards past the transducers. Furthermore, a heat exchanger connected to a thermostat was used to maintain suspension temperatures at constant levels. The resulting setup is depicted in Fig. 1.

2.2. Attenuation modeling for large particles

Over the years, a number of approaches have been taken to address the problem of modeling attenuation by suspended particulate materials. On of the models, the ECAH theory (named after their creators Epstein and Carhart [6] and Allegra and Hawley [7]) is based on the interaction of scattered, visco-inertial and thermal fields around a single particle. From the equations of motion and the balanced equations for heat and impulse, the wave equations for particle and surrounding fluid are derived. For each wave equation, scattering coefficient series are formed. The series are then derived from the conditions at the phase boundary between particle and fluid. To constitute an attenuation formulation, all higher order coefficients are dropped. Explicit equations based on this approach exist for attenuation by thermal and by the visco-inertial interaction of particle, sound wave and surrounding fluid. Additionally, an expression for the absorption of sound energy in the continuous and the dispersed phase is given. Since the ECAH theory is derived from scattering around a single particle, field interactions by neighboring particles are not correctly reflected, which hinders the application to suspensions with higher concen-