Ultrasonic Particle Sizing†

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Abstract

The measurement of particle size distributions and particle concentrations is an important task in laboratories and in technical processes that deal with suspensions. Especially the instruments designed for on-line applications in production processes mostly evaluate the interactions of light with the suspended particles, viz. extinction, scattering and diffraction. While the interaction of particles with ultrasonics is essentially the same as with light, the application of ultrasonics offers special technical advantages, which are due to the extremely broad range of wavelengths and the coherent detection of ultrasonic waves. Ultrasonic spectrometry makes possible the on-line measurement of the particle size distribution and particle concentration in suspensions at concentrations of up to 30% by volume without diluting the suspension. This paper is intended to introduce the reader to the fundamentals of ultrasonic particle sizing and to explain present efforts in this field. In the last section, some successful realizations of ultrasonic particle sizing instruments are presented.

1. Introduction

Multiphase systems, especially those consisting of solid particles dispersed in a liquid, are encountered in many technical processes. Knowledge of the dispersion state of these systems is desired in most cases, whether to evaluate product quality, to optimize the efficiency of the following processing units or to operate closed-loop process-control devices. The last-mentioned case seems to be the most challenging for the technology of measurement, since fast on-line systems have to be provided. The information on the dispersion state is basically required in terms of particle concentration and particle size distribution (PSD).

A number of highly developed instruments based on light extinction, scattering or diffraction measurement are available for laboratory use, but a successful application in on-line control is problematic for various reasons: Due to multiple-scattering problems, the solids concentration is restricted to values much lower than those encountered in typical processes. Diluting the suspension before measuring is impracticable, since it increases the total cost of the system considerably and because dilution can cause changes in the dispersion state, esp. with sensitive particles such as soluble

crystals, fragile flocs, etc. Further, the wavelength of the available light sources is restricted to a discrete value (when using lasers) or to the rather narrow range of visible light, together with the adjacent regions of ultraviolet and infrared, which spans just about a decade in terms of wavelength. As a consequence, the wavelength of radiation used with optical systems cannot be adapted to the particle sizes in order to achieve a certain particle-wave interaction behaviour.

Ultrasonic waves exhibit many parallels to light, but there are important differences in practical use:

- The frequency (and thus the wavelength) is variable over a wide range.
- Ultrasonics penetrate almost any material, making measurements in coloured and even opaque media possible. Dynamic ranges of up to 100 dB can be achieved.
- Ultrasonic receivers are coherence-selective detectors and hence have a low sensitivity for scattered and multiply scattered waves.
- The different mechanisms of particle-wave interaction allow different approaches for an ultrasonic-based measurement system.
- Since lenses, etc. are not needed, mechanical stability is a minor problem.

Overall, ultrasonics offer enough interesting properties to justify the development of ultrasonic-based

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measurement systems for the monitoring of particle size distribution and concentration. In fact, this has led to the development of commercially built on-line and in-situ PSD analysers on the base of frequency-dependent extinction measurements that cover a broad range of particle sizes and can cope with extremely high particle concentrations.

This paper is intended to introduce the reader to the fundamentals of particle size analysis by means of ultrasonic waves and to review some successful realizations of particle size analysers. First, the different mechanisms of interaction between particle and wave are surveyed to provide a fundamental understanding of ultrasonic extinction and related phenomena. Then, the history of ultrasonic particle sizing from the early beginnings to the present state is reviewed. The focus is on developments that have already gained practical relevance.

2. Particle-wave interactions¹⁾

The physical background of ultrasonic particle sizing is complicated, as a variety of different mechanisms of interaction between particles and waves are superimposed and hence interfere with each other. The interaction of ultrasonic waves with suspended particles manifests itself a) in backscattering of the ultrasonic wave, b) in an extinction of the wave and c) in an influence on the wave propagation velocity (**Figure 1**).

As is subsequently shown, the influence of suspended particles on wave propagation velocity is extremely dependent on material properties such as the fluid viscosity, particle and fluid densities and compressibilities. Hence, gaining information on the particle system by measuring the wave propagation velocity is problematic and has not yet been applied to the analysis of particle size distributions successfully. On the other hand, the velocity of sound in suspensions can be measured very precisely with an adequate technical setup, as shown below; so this approach should not be excluded from the first.

Nevertheless, this paper is focused on the discussion of extinction, which is much better investigated. In the following section, the different interaction mechanisms are discussed in detail to offer a more comprehensive understanding of ultrasonic extinction and the possibilities of applying it to particle size measurements.

At low particle concentrations, the Bouguer-Lambert-Beer Law (BLBL) is valid for the formal description of ultrasonic extinction. This law is derived from an

Fig. 1 Principles of particle detection: Backscattering, extinction, phase velocity.

intensity balance for an infinitesimally thin layer of suspension:

$$dI = -I\alpha_{Ext,DS}\Delta z \quad (1)$$

where $\alpha_{Ext,DS}$ is the extinction coefficient of the disperse system. By integration, this leads to an expression for the transmission T

$$T = I/I_0 = \exp\{-\alpha_{Ext,DS}\Delta z\} \quad (2)$$

and the extinction E

$$E = -\ln I/I_0 = \alpha_{Ext,DS}\Delta z \quad (3)$$

In both cases, I_0 and I are the radiation intensities transmitted in the absence and presence of particles, respectively, and Δz is the pathlength.

From the physical point of view, it is most appropriate to express the particle concentration in terms of the particle projectional area per unit fluid volume:

$$C_{PA} = \frac{\text{particle projectional area}}{\text{suspension volume}} \quad (4)$$

and to describe the intensity of particle-wave interaction in terms of the extinction efficiency K :

$$K = \frac{\text{extinction cross-section}}{\text{particle projectional area}} \quad (5)$$

With that, the extinction coefficient of the disperse system can be interpreted as the product of the particle extinction efficiency with the particle projectional area concentration:

$$\alpha_{Ext,DS} = K \cdot C_{PA} \quad (6)$$

For practical purposes, it can be more useful to describe $\alpha_{Ext,DS}$ in terms of the volume-related particle extinction coefficient, α_{Ext} , and the particle volume concentration, C_V :

$$\alpha_{Ext,DS} = \alpha_{Ext} \cdot C_V \quad (7)$$

Of course, both interpretations should lead to consistent results, which is expressed by the equality:

$$\alpha_{Ext} = \frac{\text{extinction cross-section}}{\text{particle volume}} = \frac{1.5}{x} K \quad (8)$$

With α_{Ext} , the BLBL can be written as:

$$-\ln \frac{I}{I_0} = \alpha_{Ext} C_V \Delta z \quad (9)$$

As with light, the predominant parameter in ultrasonic particle-wave interaction is the relationship between particle size x and wavelength λ , conventionally expressed by the particle size parameter

$$\sigma = \frac{\pi x}{\lambda} = \frac{\pi x f}{c_F} \quad (10)$$

c_F : phase velocity in the fluid.

It can be shown that the different mechanisms of particle-wave interaction causing extinction are related to certain regimes of the particle size parameter σ . Therefore, it is useful to describe the extinction behaviour of suspended particles in terms of the extinction efficiency K as a function of σ for the entire parameter range. As a typical example of the behaviour of solid particles, **Figure 2** illustrates the extinction efficiency of glass beads suspended in water. With large particle size parameters ($\sigma > 1$), scattering is the predominant interaction mechanism and only one parameter, σ , is required to trace the extinction function in this range. With small particle size parameters (Rayleigh range, $\sigma < 1$) however, entrain-

Fig. 2 Extinction efficiency K of glass beads suspended in water as a function of the particle size parameter $\sigma^{(1)}$.

Fig. 3 Extinction coefficient α_{Ext} as a function of particle size and frequency.

ment is the dominant interaction mechanism and K possesses a pronounced dependence on both the particle size parameter and the particle size itself. Further, K may vary by several orders of magnitude depending on the particle and fluid densities and the fluid viscosity.

Values of α_{Ext} calculated with a formula by *Lamb* and *Wolters* demonstrate the dependence of extinction on the frequency f and the particle size x (**Figure 3**). With particles of $40 \mu\text{m}$, for example, extinction at 40 MHz exceeds the value at 5 MHz by two orders of magnitude. Only for high frequencies and particle sizes do the curves coincide, since K is constant in this domain.

2.1 Mechanisms of particle-wave interaction

Going into more detail, the interaction of suspended particles with ultrasonics may be understood as being an additive superposition of the three principal interaction mechanisms depicted in **Figure 4**, viz., entrainment, scattering and resonance. Driven by pressure gradients and viscous drag forces, a suspended particle will participate in the translational movement of the surrounding fluid, but with a different amplitude and with a certain phase-lag^{2,3)}. For ultrasonic frequencies below a certain transition frequency f_t , the entrainment is dominated by viscous drag forces. Particle and fluid translational amplitudes tend to be equal and the phase-lag becomes very small; therefore, this regime is referred to as the regime of perfect

Fig. 4 Mechanisms of particle-wave interaction¹⁾.

entrainment. The suspension as a whole may be regarded as a continuum with respect to ultrasonic propagation. As a consequence, ultrasonic extinction is very low, but the wave propagation velocity in the suspension, c_{Susp} , may differ considerably from the wave propagation velocity in the pure suspending fluid, c_F . An approximate value is calculated as

$$c_{Susp} = \frac{1}{\sqrt{\chi_{Susp} \rho_{Susp}}} \quad (11)$$

where

$$\begin{aligned} \chi_{Susp} &= C_V \chi_p + (1 - C_V) \chi_F \\ \rho_{Susp} &= C_V \rho_p + (1 - C_V) \rho_F \end{aligned} \quad (12)$$

Depending on particle and fluid properties, the ultrasonic velocity can be increased or reduced by the presence of particles. For solid and liquid particles, the velocity changes to be expected are comparable to the velocity changes caused by moderate temperature variations.

The transition frequency f_t itself depends on the hydrodynamic properties of the suspending fluid and on the size of the particles. According to Skudrzyk²⁾, f_t is calculated as

$$f_t = \frac{6\eta_F}{\pi \rho_F X^2} \quad (13)$$

For frequencies above f_t and particle sizes still small compared with the wavelength, the translational movement of particles is dominated by pressure gradients and the relation of particle amplitude to fluid amplitude depends only on the particle and fluid densities. This interaction regime is therefore characterized as that of partial entrainment. With particles that are large in comparison with the wavelength, entrainment is of only marginal importance as the average of the hydrodynamic forces acting on the surface of a large particle is small compared with inertia and the particle remains stationary, interacting only by scattering.

Resonances may be excited in all deformable particles. Resonances include oscillations of the particle volume (monopole resonance) and oscillations of the particle shape (dipole and multipole resonances). A special case of great practical interest is the monopole resonance of gas bubbles, which has been intensively investigated. The approximate frequency of resonance is calculated as

$$f_{Res} = \frac{1}{\pi X} \sqrt{\frac{3\chi p}{\rho_F}} \quad (14)$$

which corresponds to the particle size parameter of resonance:

$$\sigma_{Res} = \frac{1}{C_F} \sqrt{\frac{3\chi p}{\rho_F}} \quad (15)$$

For air bubbles in water at ambient pressure, values of about $\sigma_{Res} = 0.014$ are obtained. The monopole resonance of gas bubbles is combined with extremely high extinction efficiencies ($K_{Res} = 100 - 400$) and with significant changes in the wave propagation velocity^{4,5,6,7)} (**Figures 5 and 6**).

For small particle size parameters (ca. $\sigma < 0.1$), the extinction behaviour of particles can be treated analytically, including scattering and the effect of translational particle-wave interactions³⁾. The validity of the obtained equation was investigated experimentally by Wolters⁹⁾ and was found to give reasonable results in the case of rather dense particles ($\rho_p = 2.5 \text{ g/cm}^3$, $\rho_F = 1.0 \text{ g/cm}^3$), whereas in the case of a low density difference between particle and fluid, the experimental values were considerably higher than those predicted.

In the range of higher particle size parameters, more elaborate theories must be applied in order to calculate the scattering cross-sections. Based on the work of

Fig. 5 Ratio of the total scattering cross-section to geometrical cross-section K as a function of the particle size parameter σ for a bubble and for a rigid sphere⁸⁾ (schematic graph).

Fig. 6 Measured and calculated phase velocity c as a function of the frequency near resonance⁴⁾.

Fig. 7 Calculated and measured extinction efficiency function in the range of $\sigma > 1$ ¹²⁾.

Hay and Mercer¹⁰⁾ and Faran, Jr¹¹⁾, Kräuter¹²⁾ calculated the extinction efficiency function $K(\sigma)$ analytically and compared it with measured values (**Figure 7**). The two curves show good agreement, although some assumptions are inherent in the calculation: The viscosity of the fluid, absorption in the particles and heat transfer in the system have to be negligible. In other cases, where these conditions are fulfilled less strictly, rather important discrepancies may occur between calculations and experimental results.

Attempts to calculate the velocity of sound in suspensions are known for low particle size parameters ($\sigma < 1$)^{13,14)}. These theories predict a constant value of phase velocity for $\sigma < 0.001$ (according to Eq. (11), perfect entrainment) and a rise from that particle size parameter on to the end of validity around $\sigma = 1$. The exact shape of the curves depends on the material data, viz. the viscosity of the fluid and the densities and compressibilities of both the solid and the liquid phase. Experimental results¹²⁾ with particle size parameters of ca. $\sigma = 0.005$ to 0.1 confirm the theories to some degree. Measurements of the ultrasonic phase velocity in a broad range of the particle size parameter are presented in **Figure 8**. For low values of σ , the phase velocity rises with σ as predicted by the theories. In the range of medium particle size parameters, an oscillation of c can be observed that obviously passes into the value of the pure suspending fluid when σ reaches infinity. The similarity to the extinction efficiency function (**Figure 2**) in the range of high values of σ is quite remarkable.

Fig. 8 Measured phase velocity of ultrasound as a function of the particle size parameter σ for glass beads of $x = 2 \mu\text{m}$ to $450 \mu\text{m}$ suspended in water¹²⁾.

3. Particle-particle interactions; extinction in Concentrated Suspensions^{15,16)}

A major advantage of ultrasonics compared to light is the possibility of making measurements in highly

concentrated suspensions. This section deals with the phenomena due to particle-particle interactions that occur in suspensions with increasing particle concentration. The significance of this discussion is due to the fact that so far, the evaluation of ultrasonic extinction measurements is ultimately based on the Bouguer-Lambert-Beer law of extinction, while the validity of this law is limited to low particle concentration¹⁵⁾. In view of the technical possibilities to measure with particle concentrations of up to $C_V = 0.4$ and more, it seems necessary to provide a basic understanding of particle-particle interactions. The most important interparticulate interactions which are discussed here are steric interactions, dependent scattering and multiple scattering.

- The fact that particles have a certain spatial extension and act as physical entities is the cause of steric interactions. Material bodies cannot interpenetrate, and so contact forces arise upon approach of two particles. Therefore, a particle cannot occupy any given place in the system and the arrangement of particles in a concentrated suspension is not random any more; instead, steric interactions will lead to the establishment of short-range order. This effect has to be taken into account for particle concentrations exceeding $C_V = 0.05$. Steric interactions are found to be prevalent within a wide range of concentration and particle size parameters, which is of interest regarding ultrasonic extinction measurements. Therefore, much of the authors' efforts have been focused on modeling steric interactions to understand their effect on extinction^{12,15,16)}. At present, the effect of steric interactions on extinction is well understood for the range of large particle size parameters ($\pi x/\lambda > 1$) and can be quantified by an extinction equation, which remains valid up to $C_V = 0.4$, approximately. Steric interactions lead to an increase of extinction compared to the values expected from BLBL. The effect is noticeable from about $C_V = 0.05$ onwards and becomes very significant (factor of 2 and more) with higher concentrations. In the range of smaller particle size parameters, the effects from steric interactions are masked by effects from dependent scattering.
- Dependent scattering denotes scattering processes where the mechanism of particle-wave interaction itself is modified by the presence of neighbouring particles. Two or more particles form a complex with scattering properties differing from those of an arrangement of single, independent scatterers. The physical conditions for dependent scattering are given when the interparticle clearance a is in-

ferior to the wavelength of the radiation, thus allowing an intense, mutual coupling of wave fields within and in the vicinity of neighbouring particles. In general, dependent scattering leads to a decrease of extinction. The condition for the onset of dependent scattering is $a/\lambda \leq 0.3$. From that, an expression for a critical concentration $C_{V,crit}$ can be derived that exhibits a strong influence of particle size: At a given volume concentration, dependent scattering is more likely to occur with small particles.

- Multiple scattering is the most commonly known mechanism of particle-particle interaction. The mechanism of multiple scattering implies that radiation undergoes interaction with different particles consecutively, i.e. radiation scattered by a first particle is scattered by a second particle and so forth. Multiple scattering does not imply that the mechanism of interaction between particle and wave is modified in any way. When multiply scattered radiation is received, the extinction is reduced below the correct value. Since multiple scattering in random media is a statistical process, multiply scattered radiation is characterized by a distribution of optical path lengths. Compared with radiation passing directly from the emitter to the receiver, there is a loss of coherence (if there was any before) and a broadening of the angular distribution of radiation. Multiple scattering is a major problem with optical extinction systems, whereas ultrasonic receivers are very little sensitive to incoherent radiation, as long as they are much larger than the wavelength. The amplitudes of scattered (spherical) waves or of multiply scattered waves are averaged to zero over the receiver plane (see **Figure 9**). With properly designed equipment, ultrasonic extinction or velocity measurements will in general not be affected by multiple scattering, except for the very highest concentrations.

Because different physical mechanisms are responsible for the different particle-particle interactions, conditions are found where one or the other interaction mechanism is dominant. **Figure 10** gives a map of the $\pi x/\lambda - C_V$ - plane with an outline of the regions in which steric interactions and dependent scattering, respectively, are to be expected. The curves are calculated with the mentioned criterion for the onset of dependent scattering and the 5% vol. limit for steric interactions. With an ultrasonic extinction measurement setup as described in¹⁶⁾, it is possible to distinguish between the coherent and the incoherent part of the received radiation. The results of an extinction measurement in a highly concentrated suspension obtained with this instrument are presented in

Fig. 9 The (coherent) plane wave is detected, the amplitude due to the sphere-wave is averaged to zero in the receiver.

Fig. 10 Steric interactions versus dependent scattering in the $\pi x/\lambda - C_V$ plane.

Fig. 11 Coherent (I_c) and incoherent (I_r) component of the received intensity (I_r).

Figure 11. For low concentrations, the measured extinction is in accordance with the Lambert-Beer value, since the coherent part of intensity exceeds the incoherent part and steric interactions are not significant. From a concentration of approx. 10% by volume onwards, steric interactions cause progressively higher extinctions than according to BLBL. With even higher concentrations, the influence of multiple scattering becomes apparent; this is due to the fact that the coherent part of the intensity decays faster with rising concentration than the incoherent one. In the example shown here, disturbances from multiple scattering are evident from particle concentrations of approx. 15% by volume onwards; however, with an appropriate design of the measurement system, it is possible to shift this limit to volume concentrations of about 35%.

4. Technical approaches to ultrasonic particle Sizing

Technical systems of ultrasonic particle sizing have developed only quite recently, with the availability of both fast electronic circuits for the generation and detection of ultrasound and sufficiently fast computers for the deconvolution of the spectral extinction or velocity data. In the following, a short survey of technical applications will be given, with special attention to those systems which have already gained some practical importance or which have been important for the historical development.

4.1 Measuring in gas-liquid systems

One might think that gas-bubbles in liquid systems should not be subject to a powder and particle-magazine, but since these systems exhibit unique interactions with ultrasonic waves, they are interesting enough for this paper. Gas bubbles are of practical importance in the chemical and food industry, fishery, ocean navigation, and so on. As with solid particles, there are three approaches to bubble detection^{8,17)} (**Figure 1**):

- A very effective scattering of sound when the bubble is insonified at or near its natural frequency. Back-scattering at variable frequency can be measured to determine the number of bubbles.
- The absorption and scattering causes extremely high attenuation of the sound wave, especially near the frequency of resonance (see **Figure 5**). For frequencies above the resonance, the extinction efficiency of bubbles is $K = 2$, independent of the bubble size. This effect may be used for the measurement of interfacial areas in gas-liquid systems¹⁸⁾.

- The compressibility of the bubble depends on the frequency, and so the wave propagation velocity in the suspension is a function of the frequency, too. **Figure 6** shows this phenomenon. The difference between the speed of sound in the bubbly liquid and the value in the pure liquid can be used to calculate the volume concentration.

Medwin^{8,17),1977} performed in-situ experiments in sea-water with two different setups:

- In the pulse-echo technique, a short sinusoidal wave train is sent out and reflected at a steel-plate. The same transducer receives the reflected echoes. From the damping of the echoes, the scatter along the path can be obtained, the exponential attenuation of the echo patterns in sea compared to that of pure water provides the excess absorption and, finally, the time between the echoes gives information on the local speed of propagation. The latter could not be evaluated in Medwin's experiments, since the pulse-echo technique was unable to measure the dispersion of sound speed accurately enough, but the extinction cross-section and the scattering cross-section were determined as a function of the frequency. Further, the number of bubbles in a given radius increment per unit volume of water was calculated directly, assuming that only the resonating bubbles will give a significant contribution to extinction.
- No reflections are utilized with the continuous wave technique, but two hydrophones separated by a certain distance on a line in the direction of sound propagation receive the waves instead. The speed of sound can be determined very easily by comparison of the phases in the two signals. Since it is not a plain sine-wave that is used, but a composition of sine-waves with up to 32 harmonics, a more sophisticated signal interpretation including a FFT analysis has to be employed in order to determine the attenuation. The attenuation can then be used to calculate the volume fraction of bubbles in the range of $x \dots x + \Delta x$ and, from that, the bubble density distribution function.

Gazanhes, et al.^{4),1984} have done theoretical and experimental work on the behaviour of the phase velocity and the absorption in bubbly water as a function of frequency. For their experiments, they used a Gauß-distributed bubble collective with a mean diameter of 100 μm and a bubble volume concentration of up to 10^{-4} . With an ultrasonic transmitter and a receiver in the dispersion, the ultrasonic resonance spectrum was measured in the frequency range of 15 kHz to 500 kHz. The authors were able to deter-

mine the mean bubble diameter and the standard deviation of the Gauß distribution.

4.2 Measuring in solid-liquid systems

Measurement systems for bubbles are mostly operated in the range of bubble resonance frequency, which allows a rather direct determination of the bubble size spectrum. In solid-liquid systems, such pronounced particle resonance frequencies do not exist. Instead, the sizing of solid particles relies on spectral extinction measurements covering a broad frequency range, so that every particle size class runs through at least two regimes of particle-wave interaction with different slopes in the extinction efficiency function. In this way, it is guaranteed that the system of linear equations relating the spectrum of extinctions measured at various frequencies to the spectrum of particle concentrations in the various particle size classes can be solved unambiguously. In solid-liquid systems, distinct changes in the slope occur around particle size parameters of $\sigma \approx 10^{-4} \dots 10^{-2}$ (transition from total to partial entrainment, Eq. (13)) and around $\sigma = 1 \dots 10$, and both regimes are utilized for measurements. Hence it is useful to distinguish between low-frequency and high-frequency approaches to ultrasonic particle sizing, owing to the specific advantages and problems of the respective regimes:

- $\sigma \ll 1$:
 - Low frequencies are easier to handle
 - The low extinctions associated with low particle size parameters make measurements in highly concentrated suspensions possible
- $\sigma \geq 1$:
 - Dependent scattering effects are almost non-existent
 - Material properties have less influence on the extinction efficiency than in the range $\sigma \ll 1$
 - Gas bubbles in the suspension do not necessarily cause measurement errors; their effect can be eliminated

4.2.1 Low-frequency approaches

Urlick^{19),1948}, and others investigated the extinction of ultrasonics at a single, relatively low frequency. The author found a definite relation between particle concentration and measured extinction up to high concentrations, and so the applications mainly proposed were monitoring of the solids concentration, with the other phase properties and the particle size distribution being constant.

Fritsche^{20),1968} measured the absorption coefficient of ultrasound in industrial suspensions of $(\text{NH}_4)_2\text{SO}_4$

crystals at 0.8 MHz and 4 MHz with crystals from about $10\mu\text{m}$ to $1000\mu\text{m}$. His goal was to determine the crystal concentration. The author found the absorption coefficient to rise linearly with concentration, the gradient depending on the frequency and particle size. He concluded that ultrasonic attenuation measurements are a suitable means of concentration determination in crystal suspensions, as long as the particle size distribution remains constant during operation so that calibration excludes all effects of particle size on absorption.

Two different frequencies, transmitted and received by two separate pairs of transducers, are used with the ARMCO – Autometrics PSM 100 which is based on a patent by *Cushman, et.al.*^{21),1973}. A change of particle concentration will lead to changes of the extinction which are proportional for both frequencies, while a change of the particle size distribution causes different changes in the extinction at the two frequencies. The concentration of the suspension is then calculated from the sum of the measured extinctions, their difference provides one single point of the particle size distribution curve. For this, the PSM technique can only be applied when the PSD is monomodal, of a known type and uniquely defined by a single point. Results obtained with the PSM are published in²²⁾. Detailed technical data on the instrument are not known, but it seems evident that the system operates in a frequency range where extinction is dominated by the mechanism of entrainment ($x \ll \lambda$). In this range, the analysis relies very much on exact information on the material data of particles and fluid (density, compressibility and viscosity). Further, air bubbles in the suspension lead to substantial changes in attenuation. The problem can be solved by removing the air with a device that utilizes a combination of centrifugal force and reduced pressure, but this increases the cost of the overall system considerably. The PSM is capable of measuring distributions with mean sizes ranging from $40\mu\text{m}$ to $100\mu\text{m}$ at solids concentrations of up to ca. 25% by volume²²⁾. The rather high concentration limit is due to the low frequencies used and the low extinction associated with these. The instrument is employed in the control of grinding circuits in the preparation of minerals.

*Alba and Herbst*²³⁾ developed the PROASSIST SDE-21 as an extension to the PSM. Five frequencies in the range of 1 MHz to 6 MHz are used to give an estimate of the particle size distribution in 5 intervals with an update every 30 seconds. Just as with the PSM, the SDE-21 relies on an expression for the attenuation $\alpha(x,f)$ in a monosized dispersion with

particle size parameters of $\sigma \leq 1$ ²²⁾. Assuming the total extinction to be a weighted average of the attenuations contributed by each particle size fraction, the Lambert-Beer equation is taken in a discretized, integral form for the mathematical deconvolution of the measured extinctions. The measurable range of mean sizes is equal to that of the PSM; in the case of both, the electronics and transducer configuration do not allow measurements over 10 MHz. The simple mathematical model employed is only approximately valid and empirical data are still required to provide accurate size information. Even though the SDE-21 system estimates five fractions, it cannot handle broad or narrow distributions due to the inversion mathematics it employs for deconvolution.²²⁾

Pfau^{24),1974} investigated the efficiency of flocculation with a Doppler system, measuring both the frequency shift and the intensity of backscattered ultrasonic waves. He found that either the mean particle size or the particle concentration in a suspension can be determined from the measured intensity, as long as one of these two parameters is known and constant. Experiments with 5 MHz ultrasonics in suspensions of particles ranging from about $x = 20\mu\text{m}$ to $80\mu\text{m}$ ($\sigma < 1$) and constant concentrations of 1% vol. showed a linear rise of the received intensity with particle size. The opposite case (constant particle size and variable concentration) brought linear results, too.

Boxman, Scott and Jochen^{25),1995} report on a prototype in-line ultrasonic cell for measuring particle size distributions in dilute ($< 4\%$ vol.) slurries of submicron particles. In their effort to develop a high-concentration ultrasonic PSD-analyser, the authors are working on a model of ultrasonic attenuation that can cope with the effects occurring at higher concentrations, whereat they focus on multiple scattering. Further, measurements of attenuation spectra in the frequency range of 2 MHz to 50 MHz at concentrations of 0.5% to 40% (vol.) were performed that exhibit the expected deviations from linearity from approx. 5% onwards. Attempts to extract PSD information by inverting the measured ultrasonic spectra were only partially successful, as the authors report. They suggest to resign from deconvoluting and to consider the attenuation spectrum as being a good representation of the actual sample.

4.2.2 High-frequency approaches

Horak^{26),1977} was interested in the physiological properties of blood, e.g. the aggregation of red blood cells as a function of the stirring rate in a sample. He has performed measurements of the ultrasonic

attenuation spectra within a frequency range of 4 MHz to 50 MHz in human blood and in suspensions of polystyrene spheres in water. Due to the relatively high frequencies and particle sizes of up to 300 μm , it was possible to investigate the attenuation behaviour with particle size parameters of $\sigma \gg 1$. The slope of the change in attenuation around $\sigma \approx 1$ turned out to be a measure of the variation of the particle diameters in the suspension. The spread of the PSD could thus be determined by this slope, and the average particle size by the frequency at which the change in attenuation took place. The author has suggested using attenuation measurements to extend Doppler systems as they are used in medicine. These devices analyse, for example, the flow rate of blood in a vessel by evaluating the frequency shift of reflected ultrasonic waves; information on the aggregation of the red blood cells was obtained by attenuation measurement.

Riebel and Löffler¹⁾¹⁹⁸⁹, have developed an on-line particle size and concentration monitor based on the measurement of ultrasonic extinction in suspensions at M different frequencies f_i in a relatively broad range from ca. 0.5 MHz to 100 MHz. In spite of the technological difficulties, high-frequency ultrasonics were chosen because extinction is less dependent on particle shape and material properties in the domain of high particle size parameters ($\sigma > 1$). Contrary to pulse-echo systems, for example, the suspension is insonified continuously; therefore only very low intensities are needed and sensitive particle structures like agglomerates are not destroyed so easily.

For the mathematical deconvolution of the measurements, the Lambert-Beer equation for the extinction from a particle size distribution

$$-\ln \left(\frac{I}{I_0} \right)_{f_i} = \Delta z C_{PA} \int_{x_{\min}}^{x_{\max}} K(f_i, x) q_2(x) dx \quad (16)$$

is approximated by the discretized version

$$-\frac{1}{\Delta z} \ln \left(\frac{I}{I_0} \right)_{f_i} = C_{PA} \sum_j K(f_i, x_j) q_{2,j} \Delta x_j \quad (17)$$

for discrete particle size intervals j , $j = 1 \dots N$. With M different measuring frequencies f_i , a set of linear equations is obtained which is expressed as

$$m = K g \quad (18)$$

where m = vector of measured extinction values,

$$-\frac{1}{\Delta z} \ln \left(\frac{I}{I_0} \right);$$

g = vector of particle size distribution, $C_{PA} q_2(x) dx$;
 K = matrix of extinction coefficients.

The values of $K(f_i, x_j)$ are taken from measured $K(\sigma)$ functions and are stored in the system software. As with most indirect methods of measurement, for example laser diffraction spectroscopy²⁷⁾, the results obtained from a direct inversion of Eq. (18):

$$g = K^{-1} m \quad (19)$$

tend to be unstable and very sensitive to minute measurement errors; special inversion algorithms have to be applied. In the course of the authors' work, several different deconvolution algorithms were tested by means of experiments and computer simulations. Their performance can be evaluated in terms of the behaviour with very narrow, very broad or polymodal particle size distributions, very high concentrations, different solid/liquid systems, and so on. The algorithm implemented finally can be regarded as a successful compromise, since the system can handle any given PSD.

Kräuter and Riebel²⁸⁾ have recently introduced an interesting alternative to the usual deconvolution techniques. They employed a neural network to estimate particle size distributions from measured extinctions. No knowledge whatsoever of particle-wave interaction, etc. is required, but the network has to be trained with data from measurements or from reliable simulations. Dependent on the desired output quality, the number of necessary training steps can exceed 2000. Experiments with monomodal particle size distributions and concentrations of up to 20% by volume brought results comparable to those of standard inversion methods.

A commercial version of the above-mentioned development is the OPUS (On-line Particle size analysis by Ultrasonic Spectrometry) system offered by SYMPA-TEC, which has found applications for the on-line-control of solid-liquid systems (emulsion polymerization, crystallization) in the chemical industries. At the present stage of development, the OPUS allows measurements of concentrations and particle size distributions of any type in a range from ca. 2 μm to 3000 μm at concentrations of up to 25% – 30% by volume. As a result of the high frequencies used with the system, gas bubbles appear as particles in the uppermost particle size interval, and so the influence of gas bubbles in the suspension can be eliminated

quite easily. A complete measurement takes approx. 20 seconds. **Figures 12 to 14** show some results obtained with the OPUS. **Figure 12** gives an impression of the ability of ultrasonic particle sizing to cope with extremely high particle concentrations and low density contrasts ($\rho_{\text{polystyrene}} = 1.06 \text{ g/cm}^3$). The variance in the measured PSDs is due to the high-concentration effects discussed in section 3, viz. steric interactions and dependent scattering. Measurements with smaller and more dense particles yield no concentration dependence at all (**Figure 13**). For comparison purposes, a measurement obtained with laser diffraction analysis is shown here. The deviation can be explained by the different equivalent diameters the two systems give; equal results can only be expected for ideal spheres. Very small particles are not the preferred domain of the OPUS, but particles as small as $1 \mu\text{m}$ in diameter can be analysed without problem (**Figure 14**).

Recently, *Pendse and Sharma*^{29),1993} introduced the Pen Kem 8000, an ultrasonic PSD estimator designed especially for slurries of inorganic, submicron particles. Similar to the OPUS, the system measures ultrasonic attenuation spectra at several frequencies in the range of 1 MHz to 100 MHz, but, as far as it is evident from the author's contribution, the mathematical deconvolution technique and the physical model of particle-wave interaction employed differ considerably. Only the viscous energy dissipation due to the relative motion of particle and fluid is taken into account, restricting the validity of the model to low particle size parameters and high density contrasts ($\Delta\rho/\rho_F \gg 1$). The authors report on results from measurements in titanium dioxide slurries with concentrations of 3.5% and 42% by volume and particle sizes from ca. $0.1 \mu\text{m}$ to $10 \mu\text{m}$. A PSD estimate takes about 10 minutes.

5. Conclusion

Of the various possibilities to detect and size particles by means of ultrasound, the extinction methods appear to be the most feasible from the technical point of view. A variety of technical solutions, ranging from ultrasonic extinction measurements at a single frequency and measurements at two frequencies to spectrometric extinction measurements with a multitude of measuring frequencies has developed during the past two decades. While the technical details differ greatly, a common feature of ultrasonic extinction measurement appears to be the ability to

Fig. 12 Particle size distributions of polystyrene beads ($\rho_{\text{polystyrene}} = 1.06 \text{ g/cm}^3$) in water measured with the OPUS. The effect of steric interactions and dependent scattering causes concentration-dependent deviations in a certain particle size range.

Fig. 13 Measured PSDs for suspensions of quartz sand in water. The deviations from laser diffraction measurement are due to the different equivalent diameters with the two systems.

Fig. 14 PSD of zeolite crystals ranging from $x = 1 \mu\text{m}$ to $100 \mu\text{m}$ measured with the OPUS and with laser diffraction for comparison.

measure in highly concentrated suspensions, which offers unequalled prospects for applications in on-line measurement and process control.

Symbols

C_{PA}	particle projectional area concentration	$[m^{-1}]$
C_V	volume concentration	$[-]$
c	sonic velocity	$[m\ s^{-1}]$
f	frequency	$[s^{-1}]$
I	ultrasonic intensity	$[kg\ s^{-2}]$
K	extinction efficiency	$[-]$
p	pressure	$[kg\ m^{-1}\ s^{-2}]$
q_2	particle area density distribution	$[m^{-1}]$
x	particle diameter	$[m]$
z	length	$[m]$
α	extinction coefficient	$[m^{-1}]$
χ	ratio of specific heats of gas at constant pressure and constant volume	$[-]$
η	dynamic viscosity	$[kg\ m^{-1}\ s^{-1}]$
κ	compressibility	$[m\ s^2\ kg^{-1}]$
λ	wavelength	$[m]$
ρ	specific density	$[kg\ m^{-3}]$
σ	particle size parameter ($= \pi x/\lambda$)	$[-]$

Indices:

0	value in pure suspending fluid
DS	disperse system
Ext	extinction
F	fluid
P	particle
PA	projectional area
Res	resonant
$Scat$	scattering
$Susp$	suspension
V	volume

Operators:

Δ	discrete difference
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Author's short biography

Ulrich Riebel

Professor Ulrich Riebel received his doctoral degree in 1988 with a thesis on ultrasonic particle sizing at the University of Karlsruhe (TH), where he had worked as a research assistant of Prof. Friedrich Löffler at the Institute for Mechanical Process Engineering and Mechanics ("Institut für Mechanische Verfahrenstechnik und Mechanik"). After having worked with the Sympatec Company in 1989/90, he went back to Friedrich Löffler's institute as a senior scientist. Since July of 1994 Ulrich Riebel holds the Chair of Particle Technology at the University of Cottbus. His main research interests are particle sizing by means of ultrasound and light especially in highly concentrated suspensions, dust separation and inter-particulate forces in crystal suspensions.

Peter Khatchikian

The author studied chemical engineering at the University of Karlsruhe (TH) and received his diploma in 1994. Since July of 1994 he is research assistant of Ulrich Riebel at the University of Cottbus. Peter Khatchikian's main research interests are the development of new methods in ultrasonic particle sizing and the statistics of signals in extinction measurement.

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Udo Kräuter received his diploma in chemical engineering from the University of Karlsruhe (TH) in 1990. Since then he is research assistant of Ulrich Riebel. Udo Kräuter works on several problems that deal with ultrasonic particle sizing, e.g. wave propagation in highly concentrated suspensions. He is also interested in the statistics of transmission signals in light extinction measurement.