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Measurement and characterization of particles by acoustic methods —

Part 1:
Concepts and procedures in ultrasonic attenuation spectroscopy

Mesurage et caractérisation des particules par des méthodes acoustiques —

Partie 1: Concepts et modes opératoires en spectroscopie d'atténuation ultrasonique

Reference number
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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

International Standards are drafted in accordance with the rules given in the ISO/IEC Directives, Part 2.

The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

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ISO 20998-1 was prepared by Technical Committee ISO/TC 24, *Sieves, sieving and other sizing methods*, Subcommittee SC 4, *Sizing by methods other than sieving*.

ISO 20998 consists of the following parts, under the general title *Measurement and characterization of particles by acoustic methods*:

— *Part 1: Concepts and procedures in ultrasonic attenuation spectroscopy*

The following parts are under preparation:

— *Part 2: Guidelines for linear theory*

— *Part 3: Guidelines for non-linear theory*

Introduction

It is well known that ultrasonic spectroscopy can be used to measure particle size distribution (PSD) in colloids, dispersions, and emulsions (see [6][7][8][9]). The basic concept is to measure the frequency-dependent attenuation or velocity of the ultrasound as it passes through the sample. This attenuation includes contributions due to scattering or absorption by particles in the sample, and the size distribution and concentration of dispersed material determines the attenuation spectrum (see [10][11][12]). Once this connection is established by empirical observation or by theoretical calculations, one can in principle estimate the PSD from the ultrasonic data. Ultrasonic techniques are useful for dynamic on-line measurements in concentrated slurries and emulsions. Traditionally, such measurements have been made off-line in a quality control laboratory, and constraints imposed by the instrumentation have required the use of diluted samples. By making in-process ultrasonic measurements at full concentration, one does not risk altering the dispersion state of the sample. In addition, dynamic processes (such as flocculation, dispersion, and comminution) can be observed directly in real time (see [13]). This data can be used in process control schemes to improve both the manufacturing process and the product performance.

ISO 20998 consists of three parts:

— Part 1 introduces the terminology, concepts and procedures for measuring ultrasonic attenuation spectra;

— Part 2 provides guidelines for determining particle size information from the measured spectra for cases where the spectrum is a linear function of the particle volume fraction;

— Part 3 addresses the determination of particle size for cases where the spectrum is not a linear function of volume fraction.

Measurement and characterization of particles by acoustic methods —

Part 1: Concepts and procedures in ultrasonic attenuation spectroscopy

1 Scope

This part of ISO 20998 describes ultrasonic methods for determining the size distributions of one or more material phases dispersed in a liquid. Colloids, dispersions, slurries and emulsions are within the scope of this part of ISO 20998. The typical particle size for such analysis ranges from 10 nm to 3 mm, although particles outside this range have also been successfully measured. Measurements can be made for concentrations of the dispersed phase ranging from 0,1 % by volume up to 50 % or more by volume, depending on the density contrast between the continuous and the dispersed phases. These methods can be used to monitor dynamic changes in the size distribution, including agglomeration or flocculation in concentrated systems.

2 Terms and definitions

For the purposes of this document, the following terms apply:

2.1
absorption
direct reduction of incident ultrasonic energy by means other than scattering

2.2
attenuation
extinction
total reduction of incident ultrasonic energy, including both scattering and absorption.

NOTE The recommended measurement unit is the decibel (dB), which is defined as 10 times the common (base 10) logarithm of the ratio of incident intensity to transmitted intensity, or equivalently 20 times the common logarithm of the ratio of incident amplitude to transmitted amplitude. The neper (Np) is a permitted alternative measurement unit based on the natural logarithm, rather than the common logarithm. The conversion factor is 1 Np = 8,686 dB.

2.3
attenuation coefficient
extinction coefficient
attenuation (extinction) per unit length of ultrasonic propagation through a material, measured in units of dB/cm or Np/cm.

NOTE Attenuation coefficients are sometimes scaled by frequency, or frequency-squared, to identify the dominant attenuation mechanism. For clarity, in this part of ISO 20998, only the attenuation per unit length (in dB/cm) is considered.

2.4
attenuation spectrum
attenuation coefficient measured as a function of frequency

2.5
bandwidth
range of frequencies contained in an ultrasonic signal, typically measured as the frequency difference between the -3 dB points on a spectrum analyser

2.6
broadband
characterized as having a bandwidth that is equal to at least half of the centre frequency

2.7
digitization
act of generating a digital (quantized) representation of a continuous signal

NOTE The number of bits determines the resolution (fidelity), and the sampling rate determines the bandwidth (Nyquist criterion).

2.8
excess attenuation
incremental attenuation caused by the presence of particles in the continuous phase

2.9
Fourier transform
mathematical transform that converts a time-varying signal into its frequency components, which is often implemented in computers as a Fast Fourier Transform (FFT) algorithm

2.10
interference
wave phenomenon of cancellation or enhancement observed when two or more waves overlap

2.11
intrinsic response
frequency-dependent response of the ultrasonic spectrometer itself

NOTE This is not to be confused with the intrinsic absorption of the sample component materials.

2.12
path length
distance traversed by the ultrasonic wave between the emitting transducer and the receiver

2.13
pulse
wave of sufficiently short duration to contain broadband Fourier components

2.14
reflection
return of an ultrasonic wave at an interface or surface

2.15
scattering
removal of ultrasonic energy from the incident wave by redirection

2.16
spectrum
frequency components of a signal, typically arranged as magnitude versus frequency

2.17
tone burst
short duration of a few cycles of a sinusoidal wave

NOTE Typically, a tone burst consists of 5 to 10 cycles of a sinusoidal wave.

2.18
transducer
device for generating ultrasound from an electrical signal or vice versa

NOTE Piezoelectric devices are commonly used for this purpose.

2.19
transmission
passage of ultrasound through a sample

2.20
transmission spectrum
transmission value measured as a function of frequency

2.21
transmission value
amplitude of an ultrasonic signal (or a component thereof) that has been transmitted through a sample, measured in volts or arbitrary units

2.22
ultrasound
high frequency (over 20 kHz) sound waves which propagate through fluids and solids

NOTE The range employed in particle characterization is typically 100 kHz to 100 MHz.

2.23
wave
fluctuation, e.g. pressure, shear or thermal, which propagates through a physical medium

2.24
waveform
shape of the wave when seen on an oscilloscope or digitized display

2.25
wavelength
length of a wave, determined by the distance between corresponding points on successive waves

3 Sampling and reference materials

3.1 Sampling considerations

3.1.1 Dry powders

It is necessary to disperse a dry powder in a liquid before measuring the ultrasonic attenuation spectrum. A representative sample of the powder shall be used in the preparation of the liquid dispersion. It is recommended that sampling procedures be carried out in accordance with ISO 14488. Dispersion of the powder should be carried out in accordance with ISO 14887.

3.1.2 Suspensions and slurries

The apparent particle size in flocculated or poorly-dispersed systems changes as a function of the applied shear stress. Therefore, unless floc size or quality of a suspension is to be measured, it is recommended that suspension and slurries be mixed thoroughly before a sample is withdrawn for ultrasonic analysis. The stability of the suspension impacts the results.

3.1.3 Emulsions

Many phenomena affect the homogeneity of emulsions, including creaming, droplet coalescence and phase separation. These changes affect the observed ultrasonic attenuation spectrum. If the initial droplet size distribution is to be measured for unstable emulsions, it is recommended that the sample be prepared immediately before the measurement.

3.2 Reference materials

3.2.1 Reference liquid

The use of a reference liquid is required in order to verify correct operation of the ultrasonic spectrometer itself. De-gassed clean water at ambient temperature has a relatively low attenuation coefficient (see [14]). Water is therefore recommended as a reference liquid for determining the intrinsic response of the spectrometer. A procedure for de-gassing water is given in IEC 62127-1.

3.2.2 Reference sample

The use of a reference sample is recommended to verify the correct estimation of particle size distribution from the observed attenuation spectrum, but no reference material has yet been identified for general use. The user should identify a well-characterized and stable material as a standard sample for monitoring variability in the size distribution results.

4 Methods

4.1 Principles

As ultrasound passes through a suspension, slurry, colloid or emulsion, it is scattered and absorbed by the discrete phase, with the result that the intensity of the transmitted sound is diminished. The attenuation coefficient is a function of ultrasonic frequency and depends on the composition and physical state of the particulate system. The measurement of the attenuation spectrum can be used to estimate the particle size distribution and concentration. The necessary apparatus is described in 4.2.

The total measured attenuation is due to the intrinsic absorption of the continuous liquid phase, the intrinsic absorption of the dispersed phase(s), thermal losses, viscous losses and scattering losses (see [6][7]). The relative importance of these loss mechanisms depends on the material system. A theoretical or empirical model of these mechanisms can be used to convert the observed data into an estimate of the particle size or particle size distribution. There is no single general procedure for determining particle size information from the attenuation spectrum. Different models and procedures are used depending on the application and nature of the sample, as described in ISO 20998-2.

The attenuation spectrum can be measured as long as the signal-to-noise ratio is adequate. However, an *a priori* theoretical model may not exist due to lack of knowledge about the dispersed or continuous phases. In cases where there is no suitable theoretical or empirical model available to describe the interaction of ultrasound with the system of interest, the attenuation spectrum can still be used to infer relative changes in particle size (see [13]).

A variety of techniques (see Annex A) have been used to measure ultrasonic spectra. Some of these methods have been implemented in laboratory instruments and some have been used in industrial applications. Ultrasonic spectroscopy has been used to measure particle size in a variety of material systems. Example applications are listed in Annex B.

4.2 Apparatus

4.2.1 General

As a minimum, the spectrometer consists of an excitation source, one or more ultrasonic transducers, a sample cell (or flow cell, in the case of in-process instruments), a preamplifier and a means for acquiring the signal. Each of these components shall be tailored to fit the particular needs of the implementation.

4.2.2 Excitation source

The excitation source produces the electrical signal that is converted by the transducer into ultrasonic waves. This circuit determines the frequency content of the resulting ultrasonic signal. This source may produce a continuous wave at a single frequency, a frequency sweep, a set of tone bursts (which may be at various frequencies), step pulses or broadband pulses. The frequency response (bandwidth) and electrical impedance of the source should be matched to those of the transducer. The output signal level can range from a few volts up to a few hundred volts, depending on the application and transducer type.

4.2.3 Transducers

Ultrasonic transducers convert the electrical signal from the excitation source into ultrasonic waves. The active element within the transducer is typically made of a piezoelectric material (such as barium titanate) or a piezoelectric polymer film (such as PVDF). When excited by an electrical signal, the piezoelectric element constricts and relaxes, sending a longitudinal compression wave through the facing material and into the dispersion. An acoustic delay line or buffer plate may be attached to the front of the transducer to protect it.

The construction of the transducer affects the frequency response. If the backing material heavily damps the vibration of the element, the natural resonance will be de-tuned, giving a broadband response.

In the through-transmission method (see Annex A) a second transducer is used to detect the transmitted ultrasonic waves and convert them into electrical signals. Due to the reciprocity theorem, the receiving characteristics of a transducer are the same as the emission characteristics. In a pulse-echo method, the same transducer is used to send and receive the ultrasonic pulses. In both arrangements, alignment of the transducers is important, and typically becomes critical at frequencies of the order of 10 MHz and higher.

Alignment of the transducers is achieved by pivoting them relative to each other, so that the bandwidth and signal strength of the ultrasonic signal are at a maximum. This action in effect aligns the directional patterns of emission and reception, which can be skewed with respect to the mechanical axis of the transducers. If the alignment is not correct, destructive interference at the edge of the receiver distorts the transmitted signal.

4.2.4 Sample cell

The sample cell is used to contain the dispersion and maintain the transducers in alignment with each other. This element is optional, as some instruments are designed as a probe that is inserted directly into a process vessel; in such cases, the probe body holds the transducers.

If a sample cell is used, the dispersion sample shall be circulated or stirred in order to prevent sedimentation. An exception to this requirement can be made in the case of stable suspensions where particles do not settle.

For in-process applications, if a flow cell design is used in which the process fluid (i.e. dispersion) flows through the cell, it is important to maintain an open bore all the way from inlet to outlet, in order to prevent plugging.

4.2.5 Preamplifier

The preamplifier is an optional element that boosts the relatively weak signal detected by the receiving transducer. Typically, this element will add 20 dB to 60 dB of gain to the signal. The bandwidth and input impedance of the preamplifier shall meet the needs of the transducer.

4.2.6 Receiver (signal acquisition)

This required element measures the strength of the detected transmission signal. The design of the receiver depends on the ultrasonic technique. For continuous wave signals, the receiver can be a tuned circuit that measures the signal strength at that frequency. The output is connected to a low-speed digitizer to capture the transmission signal. Swept-frequency and tone-burst techniques (see Annex A) also use a tuned receiver, where the centre frequency is adjusted to track that of the ultrasonic signal. Pulsed systems use a high-speed digitizer to capture the entire waveform of the received pulse. The digitization rate is typically chosen to be several times the highest frequency component to be measured (10 MHz to 1 GHz, in common practice). The waveform is then transformed into frequency components using a Fast Fourier Transform or other discrete Fourier transform algorithm. The magnitude of the resulting complex array is the energy spectrum of the transmitted pulse.

4.3 Preparation

4.3.1 Sample requirements

In order to achieve the closest possible correspondence between ultrasonic theory and experimental observation, it is recommended that the sample be uniformly dispersed and free of air bubbles, which scatter and attenuate sound. It is recommended that the sample be of relatively low viscosity (under 0,03 Pa·s) in order to allow trapped air to escape. Enough sample shall be prepared to fill the sample cell completely. When the sample is placed in the cell, care shall be taken to ensure that no bubbles are left clinging to the transducer face.

For certain applications (such as monitoring the size of flocs), the sample need not be completely dispersed.

In some cases, it is possible to obtain useful data from systems that do contain bubbles, provided an adequate ultrasonic signal is received. Trace amounts of very small, well-dispersed bubbles contribute to the overall attenuation. It is reported that in some cases the frequency range can be chosen to distinguish the ultrasonic resonance of air bubbles from attenuation due to scattering by particles (see [6][15]). Single, large bubbles can disrupt the ultrasonic signal, but pulsed measurement systems, for example, can detect the loss of data and compensate by taking more data.

4.3.2 Sample preparation

4.3.2.1 Laboratory applications (off-line applications)

4.3.2.1.1 Suspensions

To avoid multiple scattering and other non-linear concentration effects, an initial analysis sample with a solids concentration of no more than 5 % by volume should be prepared. If this is not sufficient to give an adequate attenuation signal, it may be necessary to increase the concentration. The concentration at which non-linearity becomes an issue is determined by the density contrast between the solids and the suspending liquid, and by the ratio of the wavelength to the diameter of the particles. In general, low density solids may be prepared at a higher concentration. For dense or large particles, it is recommended that the sample be agitated or recirculated to maintain the particles in suspension. It should be noted that poorly dispersed or flocculated material will be seen as having a larger particle size, but this ability to monitor floc size has practical applications. The actual dispersion method depends upon the sample material. In the case of powders, dispersion should be carried out in accordance with ISO 14887.

Subsequent samples at higher or lower concentrations may be prepared as needed.

Slurries are suspensions with a high concentration (over 10 % by volume) of particles. Multiple scattering and particle-particle interactions lead to non-linear effects in these samples, so the direct application of linear theories may be inappropriate in this case. In such cases, useful data can still be obtained and interpreted on the basis of empirical observation. The preparation of these slurries should minimize the amount of trapped air. It is recommended that the slurry be agitated or recirculated to maintain the particles in suspension.

4.3.2.1.2 Emulsions

In the case of emulsions with droplets approximately 10 μm in diameter or larger, linear scattering theory agrees with attenuation spectroscopy measurements even at high concentrations (50 % or more by volume) of the dispersed phase (see [7][13]). For emulsions with droplets smaller than 1 μm, this agreement can break down at lower concentrations (about 20 % by volume). The preparation of the sample ought to minimize the amount of trapped air.

4.3.2.2 In-process applications and on-line applications

By definition, in-process applications of ultrasonic spectroscopy constantly examine the process stream, so there is no sample to be prepared. However, it must be ensured that a representative sample is in the measurement zone.

4.3.3 Temperature

Temperature affects the material properties of the suspension and consequently the transmission of the ultrasonic waves (see [16]). It is therefore recommended that off-line measurements be conducted while controlling the temperature of the cell. The temperature of the dispersion shall be determined and recorded before and after analysis. If the difference is more than 2 °C, then the measurement shall be repeated. In-process applications that reach a steady-state temperature are exempt from this requirement.

4.3.4 Transducer alignment

Transducers shall be mechanically aligned in order to optimize the transmission spectrum in both the time and frequency domains. In the case of an adjustable path length, the transducer alignment shall be optimized to produce the best transmission spectrum at the ends and midpoint of the transducer travel.

4.3.5 System check

Before starting a new round of measurements, laboratory spectroscopic systems shall be checked by measuring the transmission spectrum at a predetermined path length for a suitable test sample. De-gassed water may be used as a test sample. The measurement at each frequency (over the useful bandwidth of the instrument) should agree to within 5 % of previous measurements on the same test sample.

If a particle size distribution is to be estimated on the basis of the ultrasonic attenuation spectrum, the system check should include a PSD measurement of the standard sample identified in 3.2.2. The median size of the particles should agree to within 5 % of previous measurements on the same standard.

Spectroscopic systems for in-process applications shall be checked before installation in the process.

4.3.6 Background measurement

Methods that use a fixed path length (i.e. fixed transducers) require measurement of the intrinsic frequency response of the spectrometer in order to determine the attenuation. In such cases, a suitable calibration fluid with low ultrasonic attenuation (such as water) is introduced into the cell and the transmission spectrum is obtained. That spectrum is used as a background signal in subsequent calculations. The background measurement may use the transmission spectrum generated during the system check.

In-process applications shall rely upon factory calibration of the spectrometer, unless it is possible to introduce calibration fluid and measure the intrinsic response during preventive maintenance cycles.

Methods in which the acoustic path length is varied may not require a background measurement if the sample has significant attenuation compared to the background and the transducer produces a highly directional beam. However, diffraction effects due to the finite aperture of the transducer can cause significant spreading of the ultrasonic beam, with the resulting loss of ultrasonic energy at the receiver leading to an over-estimate of the attenuation coefficient. It is therefore recommended to assess beam spread by making background measurements at each intended gap and calculating the apparent attenuation coefficient as a function of

frequency. If the attenuation coefficient at any frequency in the selected range is more than 5 % of the apparent attenuation coefficient of the standard sample identified in 3.2.2, background measurements are recommended.

4.4 Measurement

4.4.1 General

The instrument should be switched on at least 10 minutes before use, in order for the electronic systems to stabilize. After addition of a sample, adequate time should be allowed for the system to reach thermal equilibrium.

The procedure for acquiring the ultrasonic spectrum depends upon whether or not the acoustic path (gap) between the transducers is held constant.

4.4.2 Measurement on systems with adjustable path length

4.4.2.1 Collection of transmission data

The measurement consists of the following steps.

a) Place the dispersion sample in the sample cell. It is necessary to cover the transducer faces completely with the sample. In laboratory applications, it is recommended to sweep the faces of the transducers to remove any bubbles.

b) Ensure adequate circulation in the cell (if necessary).

c) Adjust the path length to a predetermined gap and measure the transmission spectrum over the desired frequency range.

d) Adjust the path length to a second predetermined gap and measure the transmission spectrum over the desired frequency range.

e) (Optional) Adjust the path length to additional predetermined gaps and measure the transmission spectrum over the desired frequency range for each additional path length.

f) Convert all of the transmission values to a logarithmic scale by calculating 20 times the common logarithm of the transmission values.

g) (Recommended) Empty and clean the cell thoroughly and repeat the system check measurement.

4.4.2.2 Determination of attenuation spectrum

4.4.2.2.1 Two gaps

Calculate the attenuation coefficient at each frequency by subtracting the logarithmic transmission value measured at the wide gap from the logarithmic transmission value measured at the narrow gap and dividing that value by the difference (in cm) between the two path lengths. Similarly, the attenuation coefficients can be calculated by taking the common logarithm of the ratio of the narrow gap transmission values to the wide gap transmission values and dividing by the path length. If beam spread should be taken into account, as described in 4.3.6, the resulting value should be reduced by an amount equal to 20 times the logarithmic ratio of the background readings at those two gaps, divided by the difference in path lengths.

4.4.2.2.2 Multiple gaps

At each frequency, calculate the linear regression for the logarithmic transmission values as a function of path length (in cm). The slope of the regression is by definition the attenuation in dB/cm. If beam spread should be

taken into account, as described in 4.3.6, the transmission values used in the linear regression should be augmented by the background readings at those gaps.

4.4.3 Measurement on systems with fixed path length

4.4.3.1 Collection of transmission data

The measurement consists of the following steps.

a) Place the dispersion sample in the sample cell. It is necessary to cover the transducer faces completely with the sample. In laboratory applications, it is recommended to sweep the faces of the transducers to remove any bubbles.

b) Ensure adequate circulation in the cell (if necessary).

c) Measure the transmission spectrum over the desired frequency range.

d) Convert all of the transmission values to a logarithmic scale by calculating 20 times the common logarithm of the transmission values.

e) (Recommended) Empty and clean the cell thoroughly and repeat the system check measurement.

4.4.3.2 Determination of attenuation spectrum

Estimate the attenuation coefficient at each frequency by subtracting the logarithmic transmission value from an amount equal to 20 times the common logarithm of the background value and dividing that difference by the path length (in cm). Similarly, the attenuation coefficients can be calculated by taking the common logarithm of the ratio of the background values to the transmission values and dividing by the path length. Due to the beam spread discussed in 4.3.6, this value is slightly higher than the actual value. Applications that use a fixed path length may develop a bias in the data due to changing transducer sensitivity; it is therefore recommended that the background values be checked periodically.

4.5 Interpretation of measurement data

4.5.1 Attenuation spectrum

The attenuation spectrum is the fundamental measure of the interaction between ultrasound and the dispersion. The various ultrasonic techniques are equivalent in that they give the same attenuation spectrum for a given well-dispersed sample. At low concentrations, the attenuation at any frequency varies linearly with changes in solids concentration. In some applications, observed changes in the attenuation spectrum provide sufficient information about a sample or process; other applications require quantitative information about the particle size distribution, which has to be extracted from the attenuation spectrum.

4.5.2 Particle size distribution (optional)

The particle or droplet size distribution can be extracted from the attenuation spectrum via a model, which may be empirical or based on first principles. The use of theoretical models to extract the particle size distribution from the attenuation spectrum is introduced in Annex C. No single model can be used in all applications, but it may happen that a single application can be solved with more than one model.

5 Reporting of results

5.1 Reports for laboratory testing

The report shall include:

— a reference to ISO 20998,

— the name of the testing establishment,

— the date of the test,

— the unique report identification,

— the operator identification,

— the instrument type used,

— the measurement method,

— the test sample identification,

— the acoustic path length,

— the frequency range of operation,

— the suspending liquid, its temperature, density and viscosity,

— the dispersed phase, its concentration, density and viscosity (if a liquid),

— the dispersing agent and amount used,

— the method of dispersion of the suspension, including dispersion time,

— any other factor which could have an influence on the results,

— the attenuation spectrum, and

— (optional) the particle size distribution (PSD). If the PSD is calculated, the report ought to include a figure of merit for the fit of the model to the observed data. The values of all material properties used in the calculation shall be included.

5.2 Results for in-process and control applications

A separate written report would be impractical for in-process and control applications. However, the measurement system shall provide the following outputs to the control system or operator:

— the attenuation coefficient value (in dB/cm) at one or more frequencies;

— (optional) the median particle size (in micrometres);

— (optional) the width of the size distribution (in micrometres);

— (optional) the solids concentration as determined by the measurement.

Annex A
(informative)

Techniques

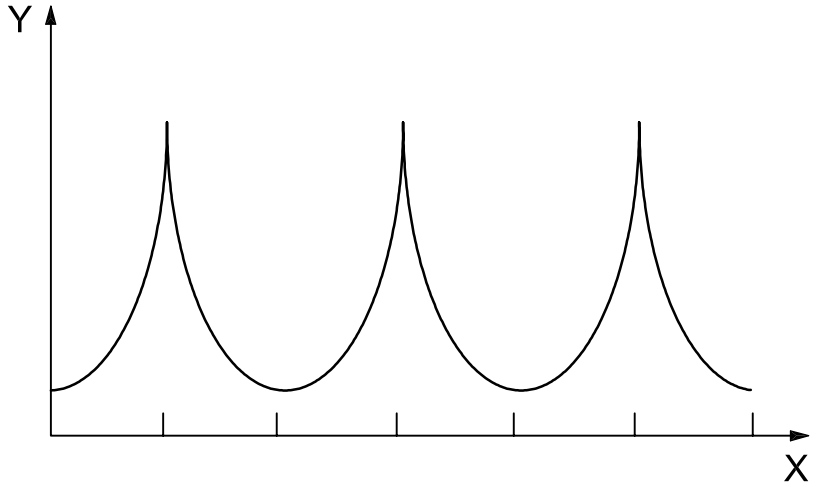
A.1 General

In order to measure the ultrasonic attenuation spectrum, it is necessary to launch ultrasonic waves into a sample and then detect the transmitted waves. Ultrasonic transducers are typically used to launch and detect the compressional ultrasonic waves, converting electrical signals into pressure fluctuations and vice versa. There are many techniques that can be used to obtain the desired spectral information. In broad terms, these may be differentiated according to the choice of excitation wave, the acoustic path length and the detection scheme. Although these techniques can provide equivalent measurements, in practice one technique may be better suited to a specific application than the others.

A.2 Excitation

A.2.1 Continuous wave

Continuous wave excitation is used for narrowband (single-frequency) transducers. The output of a radio-frequency (RF) oscillator producing a signal with a fixed frequency is applied to an ultrasonic transducer. The signal is of constant amplitude and the application period is sufficiently long that the duration can be considered infinite. The resulting acoustic signal propagates through the sample and is reflected at the opposite wall of the sample cell; thereafter the signal is repeatedly re-reflected at both ends of the sample cell. After a sufficiently long period, the transmitted acoustic signal reaches constant value. The length of the cell determines the relative phase between reflections, thereby producing varying degrees of constructive and destructive interference. The received signal varies with distance in a cyclic pattern, where the maxima occur at distances equal to a whole multiple of wavelengths. The acoustic path length must be adjusted for maximum signal at the receiving transducer, which may be the emitting transducer.



Key

X path length, mm

Y amplitude

Figure A.1 — Constructive and destructive interference for continuous wave excitation

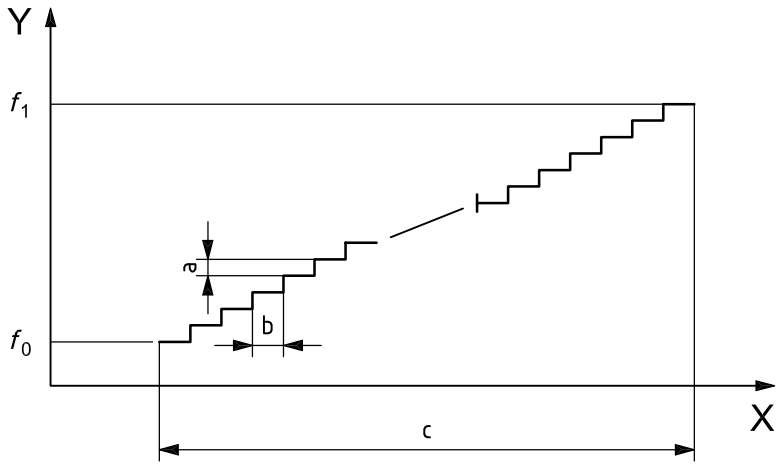
A.2.2 Frequency sweep

A.2.2.1 Continuous (analogue) frequency sweep

A voltage-controlled RF sine wave oscillator is modulated using a slow ramp waveform to produce an RF signal with a frequency increasing linearly in time. This signal is applied to the emitting ultrasonic transducer. After passing through the sample, the received ultrasonic signal is detected by the receiving transducer and can be demodulated using a phase-sensitive detector. The quadrature outputs of the detector contain amplitude and phase information of the ratio of the transmitted signal to the incident (emitted) signal. Reflections within the sample cause constructive and destructive interference, hence the spectrum is characterised by a series of peaks at the harmonic frequencies. The amplitude of these peaks provides the transmission spectrum of the sample.

A.2.2.2 Discrete (digital) frequency sweep

The frequency sweep is a pseudo continuous wave method. Each frequency is applied for a sufficiently long duration for an almost steady state condition to exist within the sample's acoustic field, at which time the magnitude and phase of the sound wave is measured.



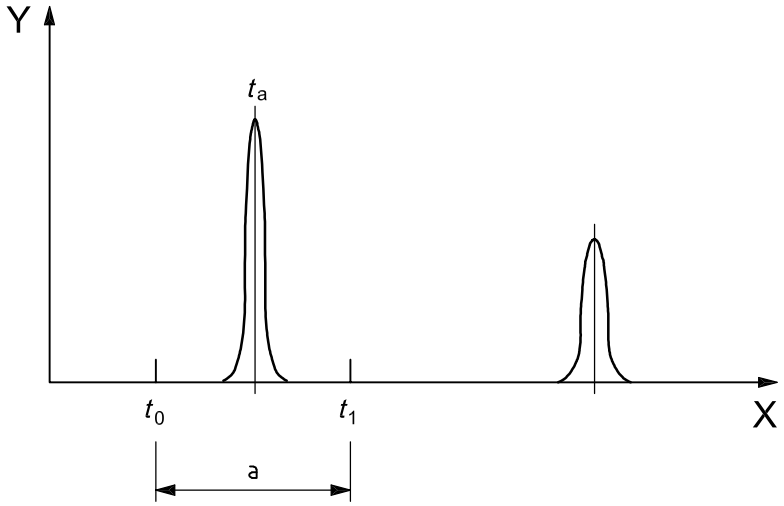
Key

X time
Y transmitted frequency
f_0 frequency start
f_1 frequency stop

a Frequency step.
b Application time.
c Sweep time.

Figure A.2 — Discrete frequency sweep

The frequency is stepped sequentially through a range of frequencies, and at the end of each application time the magnitude and phase are recorded. At the end of the sweep, the information for each frequency is combined to form a frequency spectrum. This spectrum is transformed to the time domain using an inverse Fourier transform. The magnitude of this time domain response contains a series of spikes, each representing an echo within the sample. This time domain signal is gated to isolate the first peak, which represents the direct transmission signal. Data outside this gate time is ignored; the frequency spectrum of the first peak is determined using a Fast Fourier Transform.



Key

X time

Y time domain response

t_0 start time

t_1 stop time

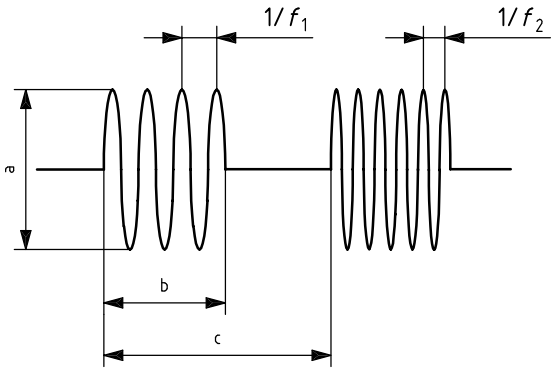
t_a first arrival time

a Search frame.

Figure A.3 — Spectrum's time domain representation used to isolate the direct transmission signal

A.2.3 Tone burst

The tone burst method is based on a discrete frequency sweep where the continuous RF signal is gated to produce a short burst. The gate controls the envelope and therefore the spectral content of the output signal. Typically, each frequency ($f_1, f_2 \ldots f_n$) is applied for a period equal to at least 5 cycles of the lowest frequency to be measured. Each burst must be shorter than the sample transit time to prevent reflections from overlapping.



a A_{pulse}.

b t_{pulse}.

c T_{repeat}.

Figure A.4 — Tone burst method of excitation

The primary tone burst signal must be detected by a receiver with adequate bandwidth. The amplitude and phase shift of the burst is measured. Successive application over a range of frequencies allows a complete spectrum to be constructed.

A.2.4 Pulse

The pulse method uses a voltage pulse of very short duration (typically 10^{-8} to 10^{-6} s) to excite a broadband transducer over a wideband frequency range. The transducer's output is a highly damped sinusoid acoustic signal. The bandwidth of the acoustic signal is inversely proportional to the width of the excitation pulse. This acoustic pulse propagates through the sample to a receiving transducer. Echoes of the original pulse are reflected at the transducer faces and other interfaces with a discontinuous acoustic impedance, such as delay lines.

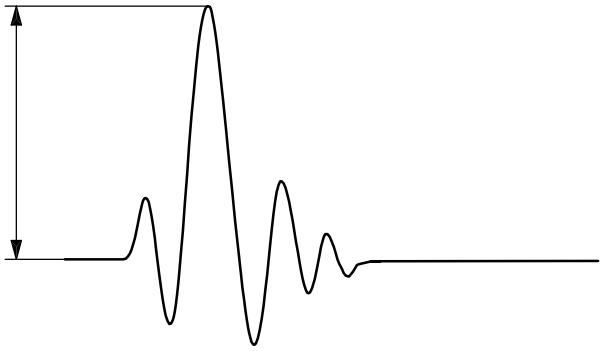


Figure A.5 — Plot of amplitude vs. time for a typical broadband pulse

The received signal is sampled by a high speed analogue-to-digital converter. The sampled signal is processed to determine the arrival time of the original pulse, and subsequent pulse echoes are ignored. This time domain data is converted to a spectrum using a Fast Fourier Transform or similar algorithm. The data from many pulses can be averaged to improve the signal-to-noise ratio. The repetition rate of the initial electrical pulse must be sufficiently low to provide time for the acoustic echoes to dissipate.

A.3 Acoustic path length

A.3.1 Fixed-path spectrometers

An accurate value of the acoustic path length is essential for determining absolute attenuation coefficients in a spectrometer where the transducer separation is fixed for the duration of the measurement. The most convenient method is to measure the propagation time of an acoustic pulse. Intrinsic time delays, such as those associated with delay lines, shall be subtracted from the observed propagation time. The sample should be temperature-controlled and well-characterised in terms of sound speed. Typically, distilled de-gassed water at 25 °C is used. The acoustic path is calculated by multiplying the propagation time by the known speed of sound in the sample.

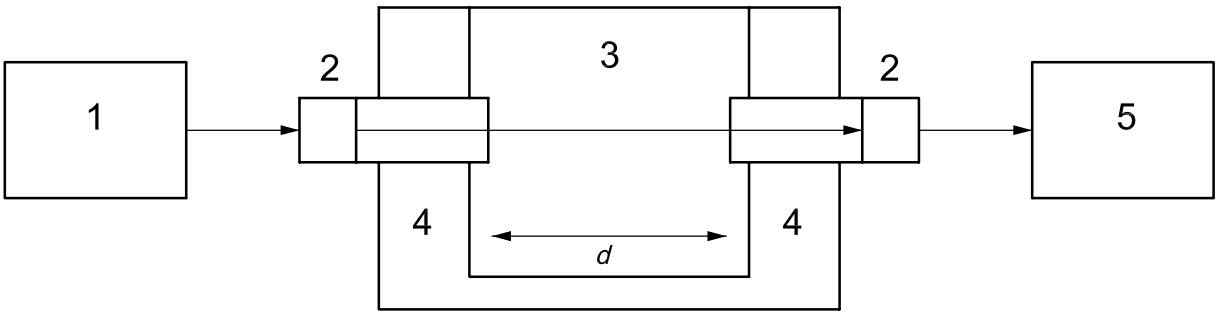
A.3.2 Variable-path spectrometers

Spectrometers with movable transducers can make a first measurement with an initial acoustic path length, then translate the transducer by a known distance, and make a second measurement with the final acoustic path length. The attenuation coefficient is determined by dividing the difference in transmitted signal by the difference in acoustic path length. A variation on this method is to measure the transmitted signal at a number of transducer separations and to compute the slope of the resulting graph.

A.4 Modes

A.4.1 Through-transmission

Through-transmission mode uses separate emitting and receiving transducers. The signal generation module is connected to the emitting transducer, sometimes via a power amplifier. The receiving transducer is connected to the detector system, usually via a preamplifier. The transducer alignment is important. If the sound beams are not co-linear, destructive interference will occur.



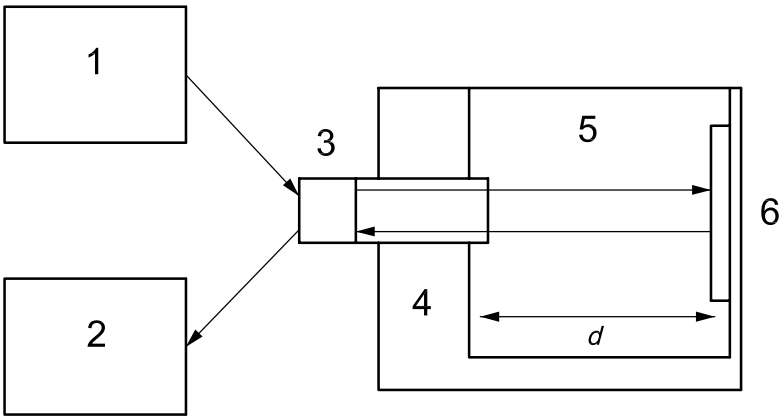
Key

1 signal generator
2 transducer
3 sample
4 delay line
5 detector
d spacing

Figure A.6 — Through-transmission mode of signal acquisition

A.4.2 Pulse echo

In the single-sided or pulse-echo mode, the same transducer is used for emitting and receiving. This mode is most often used with pulse excitation. Blanking of the excitation pulse should be used to protect the receiver if the peak voltage is in excess of approximately 10V. A wideband RF directional coupler can perform this function.



Key

1 signal generator
2 receiver
3 transducer
4 delay line
5 sample
6 reflector
d spacing

Figure A.7 — Pulse-echo mode of signal acquisition

Annex B
(informative)

Application examples

Ultrasonic attenuation spectroscopy has been used to measure the size of particles, crystals, and droplets. A few examples of such applications are listed in Table B.1. The examples shown here illustrate some of the materials that have been successfully measured. Table B.1 does not indicate the limits of applicability, and it is not meant to endorse these examples as reference materials. Details about these measurements can be found in the cited references.

Table B.1 — Examples of particle systems in which ultrasonic spectroscopy has been used to measure size

Material	Median Size µm	Concentration (vol.) %	Reference
Al_2O_3 slurry	0,3	10	[17]
TiO_2 dispersion	0,1 - 0,3	5	[18][19]
TiO_2 slurry	0,3	42	[13][20]
Urea crystals	10 - 900	6	[21]
Sunflower oil emulsion	0,5 - 10	30	[22]

Annex C
(informative)

Inversion of attenuation spectrum

C.1 Theoretical models (forward problem)

The purpose of a theoretical model is to predict the frequency-dependent attenuation of ultrasound in the sample as a function of the (known) material properties and the particle size distribution of the slurry. Predicting the attenuation in a sample with a known PSD is denoted as the “forward problem”. Once a suitable theory is identified (meaning predictions are corroborated by experiments), the problem can be inverted (reversed), so that measured ultrasonic attenuation spectra are used to predict the PSD. This inverse problem is discussed in C.2.

One widely-accepted theoretical model is the Epstein-Carhart-Allegra-Hawley (ECAH) theory, which is based on the propagation and scattering of longitudinal, shear and thermal waves in a dilute suspension or aerosol (see [11][12]). This model is linear (i.e. it is a single-scattering model) and therefore does not correctly predict acoustic properties of concentrated slurries (although it does work well for many emulsions).

Essentially, the ECAH model considers a plane pressure wave impinging on a sphere (the particle); the compression, shear and thermal waves produced can be described by three wave equations, each with its own complex wave number. The waves are then expanded in partial waves, with separate wave functions in the fluid and in the particle. By applying the boundary conditions at the surface of the sphere and invoking the orthogonality of the basis functions used in the expansion, a set of six coupled equations in six unknowns are obtained for each term in the partial wave expansion. In addition to particle size, the general ECAH model uses sixteen physical properties (eight for the liquid and eight for the particles), many of which are temperature-dependent. These properties are the sound speed, density, shear viscosity, thermal conductivity, heat capacity, heat capacity ratio, intrinsic sound attenuation and thermal expansion. Fortunately, the great majority of the real systems do not require the complete set of input parameters. For example, rigid particles under 1 μm in size (e.g. silica, alumina, rutile, other oxides, pigments, etc.) could be characterized by density alone, because variations in the thermal properties do not significantly affect the attenuation. Many emulsions, microemulsions and lattices attenuate sound independently of viscosity and density. Some other simplifications could be used in particular cases. The ECAH theory is used here as an example, but a given application may require another model, especially in the case of highly concentrated slurries where inter-particle interactions become important (see [6][7][17][18]). The volume fraction at which particle-particle interactions become important depends on the application, but in all cases it occurs when the attenuation spectrum is no longer a linear function of volume fraction.

If a suitable theoretical model is not available, empirical observations can be used to establish an approximate correlation between particle size and the attenuation spectrum. This heuristic approach shall be used with caution, because it can lead to significant uncertainty in the particle size estimate.

C.2 Recovery of PSD (inverse problem)

The inverse problem estimates model parameters (such as particle size) from observed attenuation data; it complements the forward problem where the model parameters are already known and the attenuation can be predicted. In the case of a dilute suspension, the interaction between particles is negligible. In this case, the spectrum is formed by the superposition of individual, uncorrelated events, and the problem is linear. In this case, the connection between the model, PSD and attenuation spectrum can be expressed in terms of linear algebra. Unfortunately, a direct algebraic inversion often yields results that are physically unacceptable (e.g. negative size fractions). This inverse problem is one of a general class of ill-posed problems, where the solution is not unique. Small amounts of measurement noise in the attenuation data typically lead to numerical instability in the answer. Even if the data are measured with complete accuracy, they may not convey

sufficient information. In this case, the frequency range used to observe the attenuation spectrum should be chosen with care.

A robust method of inverting ultrasonic data is to use an iterative approach (such as least-squares fitting) to minimize the difference between a calculated attenuation spectrum (based on a particular model and estimated PSD) and the measured attenuation spectrum. This method tends to provide reasonable results, even in the case of noise or incomplete data. An additional improvement in noise immunity can be achieved by imposing a connection between the individual size fractions through the assumption of a particular PSD function. Many particle systems can be described by a log-normal distribution, but other functions can also be used. The adoption of a PSD function greatly reduces the number of variables to be estimated: in the case of a single mode log-normal distribution, the only variables are concentration, median size and the width of the distribution.

A simple algorithm for inverting ultrasonic data consists of the following steps:

a) make an initial estimate of the model variables;

b) calculate the PSD from the assumed distribution function;

c) calculate the estimated spectrum predicted by the model for the PSD;

d) compare the calculated spectrum with the observed spectrum, and check for convergence;

e) if convergence is not reached, update the estimated model parameters and go to step b).

Bias in the spectral data propagates through this inversion process and is in some measure consolidated into the estimate of PSD. Possible sources of bias include:

— inadequate dispersion of the particles,

— temperature variation,

— misalignment of the transducers, and

— incorrect assessment of the intrinsic response.

Care should be taken in order to avoid these and other sources of uncertainty, since they directly affect the merit of the final solution. Bias may also originate from the mathematics used in the inversion, or from the use of an inadequate physical model.

Explicit examples of extracting particle size from attenuation spectra are given in ISO 20998-2.

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