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Measurement and characterization of particles by acoustic methods —

Part 2: Guidelines for linear theory

Caractérisation des particules par des méthodes acoustiques —

Partie 2: Théorie linéaire

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ISO copyright office
Case postale 56 • CH-1211 Geneva 20
Tel. + 41 22 749 01 11
Fax + 41 22 749 09 47
E-mail copyright@iso.org
Web www.iso.org

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Foreword

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The committee responsible for this document is ISO/TC 24, *Particle characterization including sieving*, Subcommittee SC 4, *Particle characterization.*

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*

— *Part 2: Guidelines for linear theory*

Introduction

It is well known that ultrasonic spectroscopy can be used to measure particle size distribution (PSD) in colloids, dispersions, and emulsions (References [1][2][3][4]). The basic concept is to measure the frequency-dependent attenuation or velocity of the ultrasound as it passes through the sample. The attenuation spectrum is affected by scattering or absorption of ultrasound by particles in the sample, and it is a function of the size distribution and concentration of particles (References [5][6][7]). Once this relationship is established by empirical observation or by theoretical calculations, one can estimate the PSD from the ultrasonic data. Ultrasonic techniques are useful for dynamic online measurements in concentrated slurries and emulsions.

Traditionally, such measurements have been made off-line in a quality control lab, 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 (Reference [8]). These data can be used in process control schemes to improve both the manufacturing process and the product performance.

ISO 20998 consists of two parts:

— 20998-1 introduces the terminology, concepts, and procedures for measuring ultrasonic attenuation spectra;

— 20998-2 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.

A further part addressing the determination of particle size for cases where the spectrum is not a linear function of volume fraction is planned.

Measurement and characterization of particles by acoustic methods —

Part 2: Guidelines for linear theory

1 Scope

This part of ISO 20998 describes ultrasonic attenuation spectroscopy methods for determining the size distributions of a particulate phase dispersed in a liquid at dilute concentrations, where the ultrasonic attenuation spectrum is a linear function of the particle volume fraction. In this regime, particle–particle interactions are negligible. Colloids, dilute dispersions, 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. For solid particles in suspension, size measurements can be made at concentrations typically ranging from 0,1 % volume fraction up to 5 % volume fraction, depending on the density contrast between the solid and liquid phases, the particle size, and the frequency range.

NOTE See References [9][10].

For emulsions, measurements may be made at much higher concentrations. These ultrasonic methods can be used to monitor dynamic changes in the size distribution.

While it is possible to determine the particle size distribution from either the attenuation spectrum or the phase velocity spectrum, the use of attenuation data alone is recommended. The relative variation in phase velocity due to changing particle size is small compared to the mean velocity, so it is often difficult to determine the phase velocity with a high degree of accuracy, particularly at ambient temperature. Likewise, the combined use of attenuation and velocity spectra to determine the particle size is not recommended. The presence of measurement errors (i.e. "noise") in the magnitude and phase spectra can increase the ill-posed nature of the problem and reduce the stability of the inversion.

2 Normative references

The following documents, in whole or in part, are normatively referenced in this document and are indispensable for its application. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 14488:2007, *Particulate materials — Sampling and sample splitting for the determination of particulate properties*

ISO 20998-1:2006, *Measurement and characterization of particles by acoustic methods — Part 1: Concepts and procedures in ultrasonic attenuation spectroscopy*

3 Terms and definitions

For the purposes of this document, the terms and definitions in ISO 20998-1 and the following apply.

3.1
coefficient of variation
ratio of the standard deviation to the mean value

3.2
dimensionless size parameter
representation of particle size as the product of wave number and particle radius

3.3
particle radius
one-half of the particle diameter

3.4
wave number
ratio of 2π to the wavelength

4 Symbols and abbreviated terms

A	matrix representing the linear attenuation model
A_n	coefficients of series expansion in ECAH theory
a	particle radius
c	speed of sound in liquid
C_p	specific heat at constant pressure
C_{PF}	particle projection area divided by suspension volume
CV	coefficient of variability (ratio of the standard deviation to the mean value)
E	extinction at a given frequency
ECAH	Epstein-Carhart-Allegra-Hawley (theory)
f_i	frequency
H	identity matrix
h_n	Hankel functions of the first kind
I	transmitted intensity of ultrasound
I_0	incident intensity of ultrasound
i	the imaginary number
inv()	matrix inverse operation
K	extinction efficiency (extinction cross section divided by particle projection area)
K	matrix representation of the kernel function (the ultrasonic model)
$\mathbf{K}^T$	transpose of matrix **K**
$k(f, x)$	kernel function
k_c, k_T, k_s	wave numbers of the compressional, thermal, and shear waves
ka	dimensionless size parameter
P_n	Legendre polynomials
PSD	particle size distribution

$\boldsymbol{q}$	solution vector (representation of the PSD)
$q_3(x)$	volume weighted density function of the PSD
$Q_3(x)$	volume weighted cumulative PSD
s	standard deviation
x	particle diameter
x_{10}	the 10th percentile of the cumulative PSD
x_{50}	median size (50th percentile)
x_{90}	the 90th percentile of the cumulative PSD
x_{min}	minimum particle size in a sample
x_{max}	maximum particle size in a sample
α	total ultrasonic attenuation coefficient
$\boldsymbol{\alpha}$	attenuation spectrum
$\tilde{\alpha}$	absolute attenuation coefficient divided by the frequency, $\tilde{\alpha}$ = (α/f)
α_{exc}	excess attenuation coefficient, $\alpha_{exc} = \alpha - \alpha_L$
$\alpha_{exc'}$	alternate definition of excess attenuation coefficient where $\alpha_{exc}' = \alpha - \alpha_{int}$
$\boldsymbol{\alpha}_{exp}$	measured attenuation spectrum
α_{int}	intrinsic absorption coefficient of the dispersion
α_L	attenuation coefficient of the continuous (liquid) phase
$\boldsymbol{\alpha}_{mod}$	attenuation spectrum predicted by the model, given a trial PSD
α_P	attenuation coefficient of the discontinuous (particulate) phase
α_{sc}	elastic scattering component of the attenuation coefficient
α_{th}	thermal loss component of the attenuation coefficient
α_{vis}	viscoinertial loss component of the attenuation coefficient
β_T	volume thermal expansion coefficient
Δ	error in the fit, $\Delta = \left\|\boldsymbol{\alpha}_{exp} - \boldsymbol{\alpha}_{mod}\right\|$
Δ	Tikhonov regularization factor
Δl	thickness of the suspension layer
ΔQ_2	fraction of the total projection area containing a certain particle size class
η	viscosity of the liquid
κ	thermal conductivity

λ	ultrasonic wavelength
μ	shear modulus
ρ, ρ'	density of the liquid and particle, respectively
ϕ	volume concentration of the dispersed phase
χ^2	chi-squared value
Ψ_c	compression wave
Ψ_s	shear wave
Ψ_T	thermal wave
ω	angular frequency (i.e. 2π times the frequency)

5 Mechanism of attenuation (dilute case)

5.1 Introduction

As ultrasound passes through a suspension, 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 is described in ISO 20998-1.

5.2 Excess attenuation coefficient

The total ultrasonic attenuation coefficient, α, is due to viscoinertial loss, thermal loss, elastic scattering, and the intrinsic absorption coefficient, α_{int}, of the dispersion (References [1][10]):

$$\alpha = \alpha_{vis} + \alpha_{th} + \alpha_{sc} + \alpha_{int} \tag{1}$$

The intrinsic absorption is determined by the absorption of sound in each homogenous phase of the dispersion. For pure phases, the attenuation coefficients, denoted α_L for the continuous (liquid) phase and α_P for the discontinuous (particulate) phase, are physical constants of the materials. In a dispersed system, intrinsic absorption occurs inside the particles and in the continuous phase, therefore,

$$\alpha_{int} \approx (1-\phi)\cdot\alpha_L + \phi\cdot\alpha_P \tag{2}$$

The excess attenuation coefficient is usually defined to be the difference between the total attenuation and the intrinsic absorption in pure (particle-free) liquid phase (References [4][7]):

$$\alpha_{exc} = \alpha - \alpha_L \tag{3}$$

With this definition, the excess attenuation coefficient is shown to be the incremental attenuation caused by the presence of particles in the continuous phase. Combining Formulae (1), (2), and (3), it can be seen that

$$\alpha_{exc} = \alpha_{vis} + \alpha_{th} + \alpha_{sc} + \phi\cdot(\alpha_P - \alpha_L) \tag{4}$$

The viscoinertial, thermal, and elastic scattering terms depend on particle size, but α_L and α_P do not. Thus, the excess attenuation coefficient contains a term that does not depend on size. When working with aqueous dispersions and rigid particles, this term can often be neglected, so that

$$\alpha_{exc} \approx \alpha_{vis} + \alpha_{th} + \alpha_{sc} \tag{5}$$

However, in some emulsions, the ultrasonic absorption in the oily phase can be significant. In that case, the definition of the excess attenuation coefficient given in Formula (3) may be modified as

$$\alpha_{exc'} = \alpha - \alpha_{int} \tag{6}$$

In this situation, Formula (5) is still valid. It should be noted that some authors express attenuation coefficient as a reduced quantity $\tilde{\alpha} = (\alpha / f)$, dividing the absolute attenuation coefficient by the frequency.

5.3 Specific attenuation mechanisms

5.3.1 Scattering

Ultrasonic scattering is the redirection of acoustic energy away from the incident beam, so it is elastic (no energy is absorbed). The scattering is a function of frequency and particle size.

5.3.2 Thermal losses

Thermal losses are due to temperature gradients generated near the surface of the particle as it is compressed by the acoustic wave. The resulting thermal waves radiate a short distance into the liquid and into the particle. Dissipation of acoustic energy caused by thermal losses is the dominant attenuation effect for soft colloidal particles, including emulsion droplets and latex droplets.

5.3.3 Viscoinertial losses

Viscoinertial losses are due to relative motion between the particles and the surrounding fluid. The particles oscillate with the acoustic pressure wave, but their inertia retards the phase of this motion. This effect becomes more pronounced with increasing contrast in density between the particles and the medium. As the liquid flows around the particle, the hydrodynamic drag introduces a frictional loss. Viscoinertial losses dominate the total attenuation for small rigid particles, such as oxides, pigments, and ceramics. An explicit calculation of the attenuation due to viscoinertial loss is given in Annex A for the case of rigid particles that are much smaller than the wavelength of sound in the fluid.

5.4 Linear models

5.4.1 Review

The attenuation of ultrasound in a dispersed system is caused by a variety of mechanisms (see 5.3), the significance of which depends on material properties, particle size, and sound frequency. Moreover, for some material systems, a linear relationship between sound attenuation and particle concentration can be observed up to concentrations of 20 % volume fraction or more, while for others, such a relationship exists only at low concentrations. This situation has led to a variety of models; two principal approaches may be distinguished.

The first is the scattering theory, which aims at the scattered sound field around a single particle. Based on this, the propagation of sound through the dispersed system can be calculated. By assuming independent scattering events and neglecting multiple scattering, the attenuation turns out to be linearly dependent on the particle concentration.

The fundamentals of the scattering theory were already presented by Rayleigh, but his approach ignored the energy dissipation by shear waves and thermal waves (viscoinertial and thermal losses). A well-known scattering theory is the ECAH (Epstein-Carhart-Allegra-Hawley) theory, a short introduction to

which is given in Annex B. The ECAH theory includes sound scattering as well as the viscoinertial and the thermal losses. It can be applied to homogenous, spherical particles with no limit regarding material properties, particle size, or sound frequency.

The second principal approach in modelling is to consider only the attenuation by viscoinertial and thermal losses, which is admissible in the long wavelength limit (where $x \ll \lambda$ or, equivalently, $ka \ll 1$) only. That restriction facilitates the inclusion of nonlinear concentration effects that are caused by the interaction of shear waves and/or thermal waves. Consequently, most of these theories are beyond the scope of this part of ISO 20998. However, linear solutions can be obtained in the limiting case of vanishing particle concentration ($\phi \to 0$). In general, these theories then agree with the ECAH theory (with regard to the modelled attenuation mechanism). Purely linear models are that of Reference [11] for the viscoinertial loss mechanism and that of Reference [12] for the thermal loss mechanism, both of which agree with ECAH results (Reference [7]).

The theoretical models may fail to accurately explain measured attenuation spectra since they hold true only for homogenous, spherical particles and require the knowledge of several physical parameters of the dispersed system. In such situations, semi-empirical approaches may be used that are based on the observation that for spheres we get

$$\tilde{\alpha}_{\mathrm{vis}} = \mathrm{f}(x^2 f) \,,$$

$$\tilde{\alpha}_{\mathrm{th}} = \mathrm{f}(x^2 f) \,,$$

and

$$\tilde{\alpha}_{\mathrm{sc}} = \mathrm{f}(xf) \,.$$

The application and derivation of such a semi-empirical model is described in Annex C.

5.4.2 Physical parameters

A number of physical properties affect the propagation of ultrasound in suspensions and emulsions. These properties (listed in Table 1) are included in the ECAH model described in Annex B. In most practical applications, many of these parameters are not known, and it is therefore difficult to compare theory with experimental observation directly. Fortunately, approximate models can be employed for many situations (cf. 5.3.1), which reduces the number of influential parameters. Moreover, some of these parameters only weakly affect the attenuation and, therefore, do not need to be known with high accuracy. Typical material systems are listed in Table 2 together with the material properties that most significantly affect the attenuation.

Table 1 — The complete set of properties for both particle and medium that affect the ultrasound propagation through a colloidal suspension

Dispersion medium	Dispersed particle	Units
Density	Density	$\mathrm{kg \cdot m^{-3}}$
Shear viscosity (microscopic)		$\mathrm{Pa \cdot s}$
	Shear modulus	Pa
Sound speed	Sound speed	$\mathrm{M \cdot s^{-1}}$
Absorption	Absorption	$\mathrm{Np \cdot m^{-1}}$, $\mathrm{dB \cdot m^{-1}}$
Heat capacity at constant pressure	Heat capacity at constant pressure	$\mathrm{J \cdot kg^{-1} \cdot K^{-1}}$