INTERNATIONAL STANDARD

ISO 20998-3

First edition
2017-04

Measurement and characterization of particles by acoustic methods —

Part 3:
Guidelines for non-linear theory

Mesurage et caractérisation des particules par des méthodes acoustiques —

Partie 3: Lignes directrices pour la théorie non linéaire

Reference number
ISO 20998-3:2017(E)

ISO copyright office
Ch. de Blandonnet 8 • CP 401
CH-1214 Vernier, Geneva, Switzerland
Tel. +41 22 749 01 11
Fax +41 22 749 09 47
copyright@iso.org
www.iso.org

Contents

Page

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.

The procedures used to develop this document and those intended for its further maintenance are described in the ISO/IEC Directives, Part 1. In particular the different approval criteria needed for the different types of ISO documents should be noted. This document was drafted in accordance with the editorial rules of the ISO/IEC Directives, Part 2 (see www.iso.org/directives).

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. ISO shall not be held responsible for identifying any or all such patent rights. Details of any patent rights identified during the development of the document will be in the Introduction and/or on the ISO list of patent declarations received (see www.iso.org/patents).

Any trade name used in this document is information given for the convenience of users and does not constitute an endorsement.

For an explanation on the voluntary nature of standards, the meaning of ISO specific terms and expressions related to conformity assessment, as well as information about ISO's adherence to the World Trade Organization (WTO) principles in the Technical Barriers to Trade (TBT) see the following URL: www.iso.org/iso/foreword.html .

This document was prepared by Technical Committee ISO/TC 24, *Particle characterization including sieving*, Subcommittee SC 4, *Particle characterization*.

A list of all the parts in the ISO 20998 series can be found on the ISO website

Introduction

Ultrasonic spectroscopy is widely used to measure particle size distribution (PSD) in colloids, dispersions, and emulsions[1][2][3][4]. The basic concept is to measure the frequency-dependent attenuation and/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[5][6][7]. Once this connection 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 offline 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 [8]. This data can be used in process control schemes to improve both the manufacturing process and the product performance.

Measurement and characterization of particles by acoustic methods —

Part 3:
Guidelines for non-linear theory

1 Scope

This document gives guidelines for ultrasonic attenuation spectroscopy methods for determining the size distributions of one or more material phases dispersed in a liquid at high concentrations, where the ultrasonic attenuation spectrum is not a linear function of the particle volume fraction. In this regime, particle-particle interactions are not negligible.

This document is applicable to colloids, dispersions, slurries, and emulsions. 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 about 5 % by volume to over 50 % by volume, depending on the density contrast between the continuous and the dispersed phases, the particle size, and the frequency range[9] [10]. These ultrasonic methods can be used to monitor dynamic changes in the size distribution, including agglomeration or flocculation.

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements of this document. 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*

ISO 20998-2:2013, *Measurement and characterization of particles by acoustic methods — Part 2: Guidelines for linear theory*

3 Terms and definitions

For the purposes of this document, the terms and definitions given in ISO 20998-1 and ISO 20998-2 apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

- — ISO Online browsing platform: available at http://www.iso.org/obp
- — IEC Electropedia: available at http://www.electropedia.org/

4 Symbols and abbreviated terms

For the purposes of this document, the following symbols and abbreviated terms apply.

a	particle radius
c, c'	speed of sound in the liquid and particle, respectively
C_P	specific heat at constant pressure
CV	coefficient of variation (ratio of the standard deviation to the mean value)
d	average distance between adjacent particles
dB	decibel
e	base of the natural logarithm
ECAH	Epstein-Carhart-Allegra-Hawley (theory)
f	frequency
G	real part of the effective coupling parameter S
i	the imaginary number
k	complex wavenumber
M	radius of shell in core-shell model
PSD	particle size distribution
R	imaginary part of the effective coupling parameter S
S	complex number representing the effective coupling between fluid and particle
SNR	ratio of signal level to noise level
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
$\boldsymbol{\alpha}$	attenuation spectrum
β, β'	compressibility of the liquid and particle, respectively
$\bar{\beta}$	mean compressibility of the slurry
δ_T	thermal wave skin depth
δ_V	viscous wave skin depth
η	viscosity of the liquid
κ	thermal conductivity
ρ, ρ'	density of the liquid and particle, respectively
$\bar{\rho}$	mean density of the slurry
ρ^*	mean density at the complementary concentration $(1-\phi)$

ϕ	volume concentration of the dispersed phase
ϕ_m	maximum volume concentration of the dispersed phase (maximum packing)
ϕ_NL	concentration at which the skin depth becomes equal to the interparticle distance
ω	angular frequency (i.e. 2π times the frequency)

5 Limits of applicability of linear theory

5.1 Multiple scattering

The interaction of a plane compressional sound wave with a particle generates three waves propagating outward: 1) a compressional wave; 2) a thermal wave; and 3) a viscous (transverse) wave. The thermal and viscous waves propagate only a short distance (of the order of 0,5 µm in water at 1 MHz) through the liquid.

In the linear model (discussed in ISO 20998-2), attenuation is directly proportional to particle volume concentration since only the forward compressional wave is considered as propagating beyond the region of a single isolated particle, and the effect of multiple particles is determined by the average superposition of their scattered fields.

However in the nonlinear model, the wave arriving at any particle is a combination of the incident wave together with all waves scattered by other particles. The resulting total scattered wave field is therefore a result of scattering of the incident wave by all particles and the rescattering (or multiple scattering) of already-scattered waves. All three wave modes (produced by scattering at a particle) contribute to the wave field at neighbouring particles, and can therefore be scattered by these neighbours, thereby producing compressional scattered waves as well as other modes. This effect creates a nonlinear concentration dependence of attenuation.

NOTE Multiple scattering depends on the configuration and aperture of the transducers as well as on the type of excitation signal, e.g. pulse, tone-burst, quasi-continuous, or continuous[20].

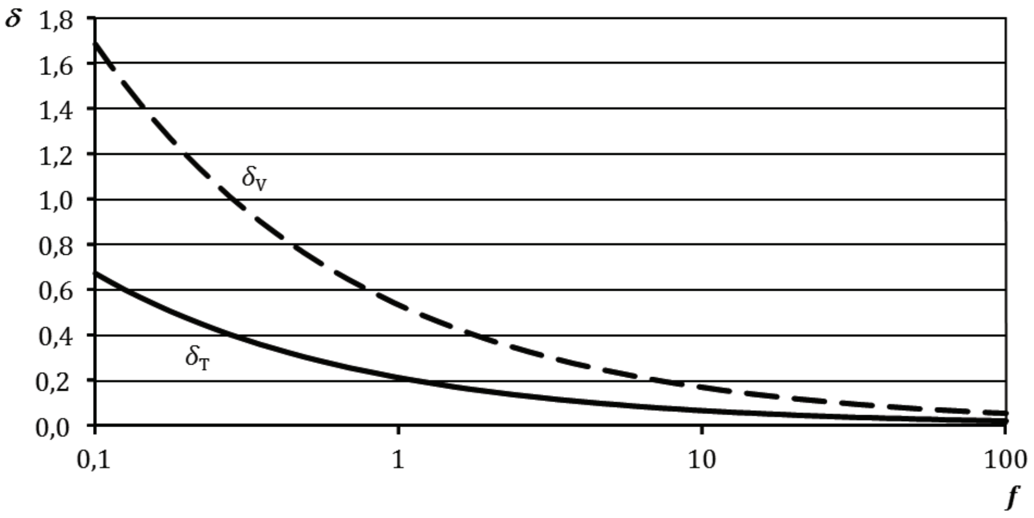
Multiple scattering models have largely considered only the multiple scattering of the compressional wave mode, neglecting the contribution of scattered thermal and shear waves to the wave field which is incident at a particle[11][12][13][14]. The second-order concentration effects obtained from these multiple scattering models are significant only where there is a substantial density difference between the phases. In many systems, the nonlinear effects due solely to compressional wave multiple scattering are small, and they can be modelled using the multiple scattering models mentioned above. Substantial nonlinear effects arise primarily because of the contributions of scattered thermal and shear waves to the incident field at any particle.

5.2 Concentration considerations

The distances over which thermal and viscous waves decrease by a factor of (1/e) are known as the thermal and viscous skin depths, respectively, which are calculated by Formulae (1) and (2). The skin depths for water are shown as a function of frequency in Figure 1.

$$\delta_\mathrm{T} = \sqrt{\frac{2\kappa}{\rho C_p \omega}} \tag{1}$$

$$\delta_\mathrm{V} = \sqrt{\frac{2\eta}{\rho \omega}} \tag{2}$$



Key

δ skin depth (μm)
f frequency (MHz)
δ_V viscous wave skin depth
δ_T thermal wave skin depth

Figure 1 — Skin depth for viscous (dashed line) and thermal (solid line) waves in water at 20 °C

At high concentrations, the interparticle spacing may become small enough that the particles can no longer be considered to be completely isolated. This effect is compounded at low frequencies, where the skin depths calculated in Formulae (1) and (2) are longer. The thermal and shear waves produced by scattering at a particle contribute significantly to the wavefield at a neighbouring particle, being rescattered to produce compressional waves (and other modes). For practical purposes, the breakdown of linear theory (or nonlinear compressional models) occurs when the viscous or thermal waves from adjacent particles overlap significantly. Quantifying the overlap in simple terms is difficult, but a standard approach is to determine the concentration when the interparticle distance is less than or equal to the skin depth. Since the viscous layer has greater thickness in most liquids, the onset of multiple scattering occurs when the interparticle distance d equals the viscous skin depth.

For a suspension of monosized spheres, the interparticle distance d is given as a function of volume concentration ϕ[15] by

$$d = x\left[\left(\frac{\phi_m}{\phi}\right)^{1/3} - 1\right] \tag{3}$$

where

ϕ_m is the maximum volume concentration based on the packing arrangement;

x is the sphere diameter.

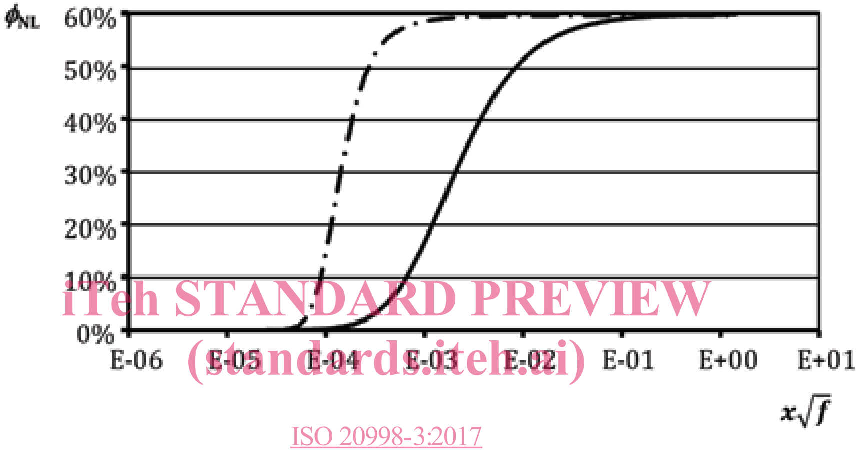
For random packing, ϕ_m typically has a value of approximately 0,6.

To determine conditions where consideration should be given to using a nonlinear model, Formulae (2) and (3) are combined to determine the concentration ϕ_{NL} at which the viscous skin depth becomes equal to the interparticle distance. The resulting Formula (4) is expressed in terms of $x\sqrt{f}$ to produce a universal function:

$$d = \phi_{\mathrm{m}}\left[\left(\sqrt{\frac{\eta}{\rho\pi}}\cdot\frac{1}{x\sqrt{f}}\right)+1\right]^{-3} \tag{4}$$

The results for aqueous slurries are shown in Figures 2 and 3; the calculations were done by assuming that ϕ_{m} = 0,6 and by substituting the viscosity and density of water.

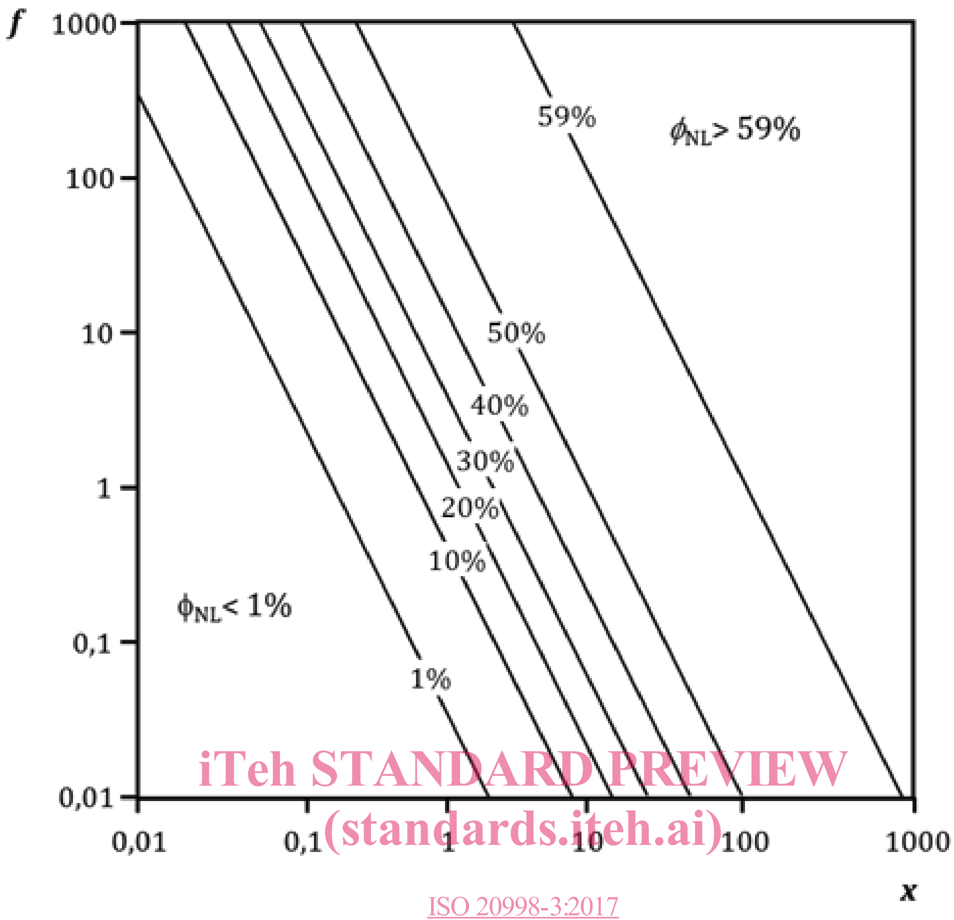
For comparison, the calculation for slurries in hexadecane (a common solvent) is included as a broken line in Figure 2. The contour plot in Figure 3 repeats the calculation for water for explicit values of frequency and particle diameter.



Key

ϕ_{NL}	volume concentration at which nonlinear effects may become evident
$x\sqrt{f}$	product of particle size and square root of ultrasonic frequency (m·Hz½)
———	water
—.—.	hexadecane

Figure 2 — Estimate of ϕ_{NL} (approximate concentration at which nonlinear effects may become evident) calculated for water and for hexadecane



Key

f frequency (MHz)

x particle diameter (mm)

Figure 3 — Contour plot of ϕ_{NL} for water, as a function of frequency and particle diameter

As a practical example, consider a hypothetical ultrasonic spectroscopy system operating at frequencies in the range of 10 MHz to 100 MHz and measuring 0,1 µm diameter particles. The results shown in Figure 3 suggest that nonlinear effects should be considered at concentrations greater than a few volume percent. On the other hand, the same system could be used to measure particle size of 10 µm particles at concentrations of 50 % or more.

NOTE 1 Formula (4) and Figures 2 and 3 are provided only as a guideline for estimating ϕ_{NL}, the concentration limit beyond which the viscous wave interacts directly with neighbouring particles.

For concentrations in excess of ϕ_{NL}, the linear theories described in ISO 20998-2 may not be adequate for estimating particle size from measurements of the ultrasonic attenuation spectrum. In that situation, theories that predict a nonlinear relationship between attenuation and volume concentration (such as those described below) may be needed.

NOTE 2 Deviation from linear theory generally becomes greater with increasing concentration, decreasing frequency, or decreasing particle size.

5.3 Steric repulsion

Additional effects arise from the development of structure in the suspension due to steric exclusion of the particles, possibly augmented by hydrodynamic interaction forces and interactions of the