Powder Technology 268 (2014) 177–190

Contents lists available at ScienceDirect

Powder Technology

journal homepage: www.elsevier.com/locate/powtec

Ultrasonic backscattering method for in-situ characterisation of concentrated dispersions

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ARTICLE INFO

Article history:
Received 14 April 2014
Received in revised form 31 July 2014
Accepted 14 August 2014
Available online 23 August 2014

Keywords:
Dispersion characterisation
Concentrated dispersion
Particle characterisation
Sound scattering
Backscattering measurement
Backscattering coefficient

ABSTRACT

An acoustic reflection measurement technique is presented, which detects and evaluates the ultrasonic waves backscattered at the particles within a liquid dispersion. The technique is based on an iterative pulse-echo method, which gathers a sufficiently high number of single reflection signals and eventually conducts a statistical analysis yielding two principal parameters of the backscattering signal: its maximum amplitude and its time-dependent decay. Theoretical considerations and experimental investigations show that maximum amplitude of the backscattering signal correlates with the averaged backscattering coefficient of the particles and that the decay rate of the backscattering signal can be attributed to sound attenuation, even though the impact of multiple scattering (i.e. non-linear concentration effects) is much more pronounced as for a transmission setup. Since the developed measurement technique is specifically designed for the in-situ characterisation of concentrated dispersion, it is necessary to adequately account for multiple scattering effects when evaluating the backscattering data. For this purpose a semi-empirical model is developed, which combines an established single particle scattering model with predictions of wave interference for defined microstructure of the dispersion. Experiments on two different types of dispersion confirm the applicability of this approach for concentrations up to 30 vol.%. The experimental results additionally show that the maximum backscattering amplitude is highly sensitive to particle size, whereas the decay rate particularly reflects the impact of particle concentration. Finally a scheme is proposed, which allows for calculating the mean particle size and the volume fraction of the dispersed phase.

1. Introduction

The characterisation of dispersions basically intends to determine both the composition and the state of the disperse system. Besides the type and mixing ratio of continuous and disperse phase, the state is of particular importance for the macroscopic dispersion properties. For instance, the size and the shape of pigment particles within a paint are decisive for the optical properties (colour, coverage) of the paint. In general, particle size and concentration, along with thermodynamic and stability issues, essentially influence the state of a dispersion. Hence, measuring particle size and concentration is compulsory for dispersion characterisation.

There is a large variety of measurement techniques for the determination of particle concentration and size or size distribution. Despite this, only few techniques facilitate the characterisation of concentrated suspensions and emulsions, e.g. for the purpose of process monitoring. Optical detection principles can usually not be employed in this case, because of the dispersions' opacity. Using ultrasound can overcome this limitation. Moreover, ultrasonic measurement techniques are in general capable to deal with field application where sampling and preparation (i.e. dilution) should be avoided (in-situ characterisation) [1].

A well-known technique, the ultrasonic attenuation spectroscopy, uses a transmission arrangement for measuring the portion of sound which is extinguished by the dispersion. Two acoustic transducers are arranged face to face with a gap between them. The frequency dependent attenuation $\alpha(f)$ is used to determine particle size distribution, for which purpose the measured attenuation is adapted to the values predicted by physical model [2,3]. For strongly attenuating media, like highly concentrated dispersions, the measuring gap has to be diminished to a few millimetres. Under these circumstances, single coarse particles or highly viscous media can plug the gap and reduce the in-situ capability of this method.

Beside the transmitted sound waves, which are evaluated within the attenuation spectroscopy, the ultrasonic waves scattered at the particles also contain information about the dispersion [4]. Taking advantage of this‘ particle-born’ sound signal, it is possible to arrange an ‘open’ measurement setup in reflection mode without any gap. In medical application this approach is used since many years to non-invasively

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http://dx.doi.org/10.1016/j.powtec.2014.08.026
0032-5910/

characterise biological tissue [5]. Also in non-destructive material testing (NDT) ultrasound reflection measurements are used to detect defects or cracks within a material matrix [6]. Acoustic backscattering measurements in ultrasound range are even used to characterise granular sediments at the sea floor [7]. Although the detection and analysis of scattered or reflected ultrasound exhibit clear advantages regarding field application, this method is not established within particle analysis yet. On the one hand, measuring the acoustic amplitude or power scattered by small particles (in micrometre range and below) is quite difficult because of the weak scattering at particles that are small compared to the wavelength of ultrasound, which applies to all particles smaller 50 μm at frequencies below 10 MHz. On the other hand, the interpretation of the measured scattering signals, based on models for sound propagation in disperse systems, is challenging because of the physical complexity due to effects of multiple scattering. There is an extensive use of ultrasonic backscattering methods within several fields of application indeed. However, within medical application acoustic backscatter is especially used for imaging purposes whereas in NDT scattering mostly originates from single inhomogeneity. Neither of both applications is aiming at the characteristics of concentrated particle systems. Hence, their specific methods of evaluating ultrasonic reflection/backscattering are not suited to the purpose of this paper.

This paper introduces an acoustic reflection technique to gather the ultrasonic waves which are reflected, respectively backscattered at the particles. The corresponding backscattering signal contains information about the scattering behaviour of the dispersion under observation. Experimental data, presented within this paper, show a high sensitivity of backscattering signal to particle size and concentration. Backscattered intensity is compared to backscattering coefficient calculated using an empirical modelling approach. Therefore, an established single particle scattering model is combined with an approach that calculates the interference of scattered waves for dispersion with defined microstructure.

2. Theory

2.1. Scattering of sound by single particles — an analytical scattering model

The existence of particles accompanies with the scattering of sound waves that are emitted into the dispersion (Appendix A). As it is well known, the scattering behaviour of a particle is determined by the size-to-wavelength ratio (between particle size $x = 2a$ and sound wavelength λ, respectively sound frequency f), expressed in form of the dimensionless wave number

$$ka = \pi \cdot x/\lambda = \pi \cdot f \cdot x/c_c, \tag{1}$$

where c_c denotes the sound velocity in the continuous phase [2,8,9]. Furthermore, scattering is affected by the acoustic contrast between particle and surrounding medium. Based on the acoustic impedance [8]

$$Z = \rho \cdot c, \tag{2}$$

which can be determined for both phases, the relation of the acoustic impedances of the disperse[1] (index d) and the continuous (index c) phase is denoted as the acoustic contrast

$$K = Z_d/Z_c. \tag{3}$$

For small values of ka ($ka < 0.1$), visco-inertial and thermal effects additionally affect scattering (ECAH and coupled phase theories) [3,10,11]. With vanishing values of ka scattering itself becomes rather weak. In general, acoustic scattering is inelastic (dissipation of energy). In particular for small values of ka, the inelastic components dominate, while the elastic scattering is rather weak. Within this work, examinations of scattering are focused on dimensionless wave numbers $ka > 0.1$. In accordance to this, only elastic scattering components will be considered.

A commonly accepted approach to describe elastic scattering was developed by Faran [12]. In contrast to the ECAH or coupled phase theories, Faran's model does not have any restrictions regarding particle size. It can be applied to solid and fluid particles because longitudinal as well as transverse waves inside the particle are considered. Anderson presented an alternative model, which is restricted to fluid particles due to the negligence of transverse waves [13,14]. Based on the material properties (acoustic contrast K) of the involved phases (particle and surrounding medium, Table 1) and the size-to-wavelength ratio ka, Faran calculated the coefficients A_n (Appendix B).

The scattering of sound can be expressed in terms of the averaged scattering coefficient $H(\theta)$, which is the averaged differential scattering cross section per unit volume (Appendix A). For a distributed particle size, determined by j size classes and a cumulative distribution function $Q(x_j)$ with

$$\sum_j \Delta Q_j = 1 \tag{4}$$

and

$$\Delta Q_j = Q\left(x_j\right) - Q\left(x_{j-1}\right), \tag{5}$$

the scattering coefficient has to multiplied with the number weights $\Delta Q_{0,j}$ in the form

$$H = c_N \cdot \sum_j \left|L\left(ka_j, \theta\right)\right|^2 \cdot \Delta Q_{0,j}, \tag{6}$$

whereat $L(ka_j, \theta)$ is the angle distribution factor comprising the coefficients A_n and the dependency on scattering angle θ (multipole expansion, Appendix A). In terms of the volume weighted size distribution $Q_3(x_j)$, Eq. (6) has to be rewritten:

$$H = \frac{6}{\pi} \cdot c_V \cdot \sum_j \left|L\left(ka_j, \theta\right)\right|^2 \cdot \frac{\Delta Q_{3,j}}{x_j^{\,3}}. \tag{7}$$

Within this formulation the particle concentration is denoted as volume concentration c_V (instead of number concentration c_N). Nevertheless, the assumption of a linear influence of particle concentration only is valid as long as scattering at one particle is not affected by the scattering at particles in its vicinity (and as long as the scattering events are incoherent).

2.2. Scattering of sound by concentrated dispersions — an empirical approach for covering non-linear effects at higher particle concentration

The acoustic scattering behaviour of dispersions, especially at higher particle concentration, substantially differs from that of single particles. With increasing particle concentration, the single particle scattering approach is no longer valid because of the decreasing average distance between the particles. Accordingly, the scattering at one particle is affected by the scattering at neighbouring particles. This fact, known as multiple scattering, has already been studied intensively for sound waves interacting with collectives of particles [2,9]. Most theoretical approaches are not valid for particle concentrations above 10 vol.%, or they are limited to specific material mixtures, due to the physical complexity of multiple scattering in dispersions [16].

For this reason an alternative approach will be used to compute the influence of particle concentration on the scattering behaviour of dispersions. Instead of considering the particles as sources of scattering, the complete dispersion as a volume with time and spatial variable

[1] For the disperse phase the acoustic impedance Z_d is calculated using the longitudinal sound velocity $c_{d,L}$ of the particle material.