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NOVEL EVALUATION ALGORITHM FOR IN-LINE DROPLET SIZE ANALYSIS OF EMULSIONS WITH ULTRASONIC EXTINCTION

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ABSTRACT

For many industrial production processes the particle size distribution and the product concentration are important actuating variables. In the context of optimising process control and process efficiency the interest in in-line particle size analyses continuously increased over the last years. Thus, especially the field of emulsions grew above the average.

Using ultrasonic extinction sensors for in-line particle size analysis allows to measure suspensions and emulsions directly within the production process almost independent of their concentration. Frequency dependent losses of ultrasonic intensity arising from particle-wave interactions are the primary result of ultrasonic extinction measurements. These measured attenuation data may be inverted for particle size distribution and product concentration.

For evaluating emulsion analyses, a completely new algorithm for predicting the interaction of emulsified droplets with ultrasonic waves was invented. Therefore, the scattering theory of [1] and [2] was extended for the requirements of absorbing particles and implemented into an approved evaluation environment.

At first, theoretical fundamentals of ultrasonic extinction are introduced with special respect to the novel evaluation algorithm. Afterwards, exemplary results of industrial emulsion applications are shown to prove the capability of the new algorithm. Latter also includes comparisons with laser diffraction measurements carried out in parallel.

1 INTRODUCTION

Today, the principle of ultrasonic extinction (USE) is widely accepted for particle size analyses within suspensions and emulsions. Especially for high concentrated particle systems, USE sensors provide distinctive advantages in comparison with optical principles. Further on, USE sensors are able to work in a broad range of environmental conditions due to their robust construction. Therefore, they offer the possibility of in-line particle size analyses, e.g. directly within a pipe or reactor.

An ultrasonic measurement results a frequency dependent attenuation signal as primary measurand. The attenuation signal results from liquid or solid particles within the measuring zone damping the ultrasonic intensity between the ultrasonic transmitter and –receiver. Attenuation signals obtained for different ultrasonic frequencies result an attenuation spectrum. For evaluating the particle size distribution (PSD) of the analysed suspension or emulsion based on the attenuation spectrum, different physical properties of the sample material have to be known yielding a material dependent matrix of related extinction cross sections (MECS). Including the MECS, a linear system of equations can be established which can be solved for the desired PSD.

This article wants to present efforts in acquiring an algorithm for the theoretical calculation of the MECS for emulsions with particle sizes above 10 µm.

2 ULTRASONIC FUNDAMENTALS

2.1 Measurement Principle

Figure 1 sketches the principle setup of an ultrasonic measurement by means of the Sympatec OPUS system. Starting from the ultrasonic transmitter, the ultrasonic waves with the intensity I_0 pass the measuring zone which contains the emulsion to be analysed. Due to absorption and scattering effects, the ultrasonic receiver detects the minor intensity I.

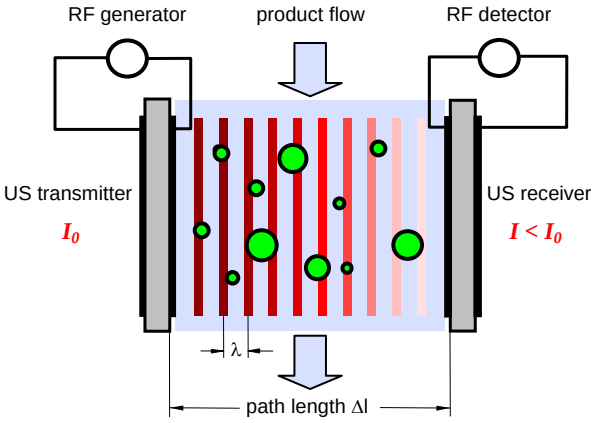

Figure 1: ultrasonic measurement principle (Sympatec OPUS)

Every USE measurement results the attenuation spectrum $A(f)$ based on a discrete number of different ultrasonic frequencies according to

$$A_i = -\ln\left(\frac{I}{I_0}\right)_{f_i} . \quad (1)$$

2.2 Mathematical Treatment

Following the LAMBERT-BEER's equation

$$\overline{A}(f) = c_{p.a.} \cdot \Delta l \cdot \overline{\overline{K}}(x, f) \cdot \overline{dQ_2(x)} , \quad (2)$$

the PSD $Q_2(x)$ can be calculated based on a measured attenuation spectrum $\overline{A}(f)$, a predicted gap width Δl and a known MECS $\overline{\overline{K}}(x, f)$. Commonly, the dependencies of the MECS are merged yielding the dimensionless size parameter σ which is calculated as

$$\sigma = \frac{\pi \cdot x}{\lambda} = \frac{\pi \cdot x \cdot f}{c} \quad (3)$$

with x particle size,
λ ultrasonic wave length,
f ultrasonic frequency,
c speed of sound.

The projection area concentration $c_{p.a.}$ in equation 2 is defined as fraction of particle projection area and emulsion volume. It is obtained as an additional result.

2.3 Matrix of related Extinction Cross Sections (MECS)

As shown in equation 2, the MECS is of prime importance for evaluating USE measurements. It describes the frequency- and particle size dependent attenuation properties of the dispersed particles.

Generally, the following mechanisms of particle-wave-interactions have to be considered:

- acoustical scattering
- thermal losses
- hydrodynamic losses (visco-inertial effect)

Due to their physical character, these attenuation mechanisms take effect within different size ranges of the dimensionless size parameter σ.

In case of $\sigma \geq 1$, the ultrasonic wave length λ is smaller than the particle size x, cf. equation 3. Therefore, it is referred to as short wavelength regime (SWR). Within the SWR, sound attenuation primary is caused by acoustical scattering.

The long wavelength regime (LWR) is defined for dimensionless size parameters $\sigma << 1$ with the wave length λ distinctively bigger than the particle size x. Within the LWR, the dominance of scattering effects is replaced by thermal and hydrodynamic losses which have to be considered nearly exclusively.

During calculation, the attenuation shares of each of the particle-wave-interactions have to be superposed for getting the total extinction cross section. Particularly, this applies for the transition range between SWR and LWR.

With regard to emulsions, an additional effect has to be considered in terms of the intrinsic absorption inside of both of the emulsion phases. The intrinsic absorption does not depend on the particle size but only on the concentration of the disperse phase.

EPSTEIN and CARHART [3] as well as ALLEGRA and HAWLEY [4] worked out the ECAH theory covering all occurring phenomena as they are known today.

3 ALGORITHM

The present work aimed at finding an algorithm for predicting the MECS for emulsions in the size range above 10 µm.

HAY and MERCER [1] gave a simplified approach to the ECAH theory by focussing on the short wavelength regime only. Thus, it becomes possible to solve the ECAH equations explicitly.

Considering scattering effects only, the elements of the MECS become

$$K_{i,j} = -\frac{4 \cdot \pi}{k_c^2} \sum_{n=0}^{\infty} (2n+1) \left| A_n(x_i, f_j) \right|^2 \quad (4)$$

with $k_C = \frac{\omega}{c}$ wave number (compressional wave),
ω angular frequency

and $A_n(x, f) = -i \cdot \sin(\eta_n(x, f)) \cdot e^{-i \cdot \eta_n(x, f)}$. (5)

The fundamental idea of describing the scattered pressure field as partial wave expansion was used by FARAN [2] to express the scattering amplitude coefficients A_n using the phase shifts η_n of the partial scattered waves.

The theory of [1] and [2] neglects the presence of intrinsic absorption. Therefore, the model was extended suitably.

In parallel, it was achieved to measure the attenuation coefficient α of both of the emulsion phases using the unmodified setup shown in figure 1. By this way, it could be ensured to consider every kind of relaxation process within the measurement frequency range between 1 MHz and 100 MHz typically. Effects of relaxation were investigated by LAMB and PINKERTON [5]. They can be considered according to

$$\alpha(f) = B \cdot f^2 + \frac{A \cdot f^2}{1 + (f / f_m)^2} \quad (6)$$

with A relaxation magnitude,
B absorption coefficient,
f_m relaxation frequency.

The new theory was integrated into an existing, approved evaluation environment.

4 PROVING THE THEORY

The verification of the algorithm was done in two steps. At first, measured attenuation spectra were compared to theoretical values calculated based on the theory. Afterwards, PSD data calculated from USE measurements were conferred to distribution data acquired with a laser diffraction (LD) instrument.

The test-emulsions were produced and characterised using the experimental setup shown in figure 2. Both liquid phases were provided in a stirred beaker ①. Using a pumped circuit ②, the emulsions were analysed with USE and LD equipment and finally returned into the beaker. The required dispersing energy was achieved by a nozzle-impact-plate ③ which had to be passed by the pumped volume flow.

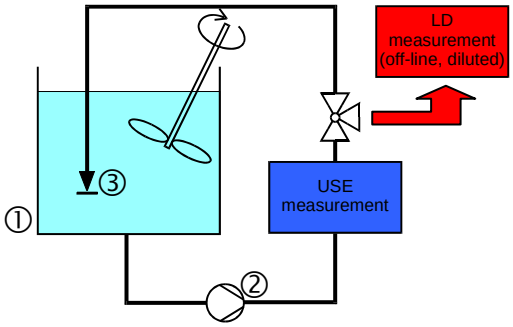

Figure 2: experimental setup for producing and characterising emulsions

All tested emulsions were based on water as continuous phase with a product concentration of 5 %vol. The measurements were performed at ambient temperature. Replacing complex systems of stabilising emulsifiers, the viscosity of the continuous phase was increased using Hydroxyethylcellulose (HEC). Thus, tendencies of coagulation could be prevented as proved during preliminary tests. In this context it turned out as a benefit of applying HEC since in contrast to emulsifying agents the acoustic properties of the emulsion are not affected.

4.1 Comparison of measured and predicted Attenuation Spectra

Using equation 2, it is possible to calculate a predicted attenuation spectrum $\overline{A}(f)$ based on a $Q_2(x)$ PSD measured with a LD instrument and a MECS acquired with the theoretical algorithm. This predicted attenuation spectrum should agree with the attenuation spectrum measured in parallel using an USE instrument.

Exemplary results for the emulsion systems

- bromobenzene in tap water
- edible oil in salt water

are given in table 1 in terms of measured absorption properties used for calculation and in figure 4 as diagram of the predicted and measured attenuation spectra.

As shown in figure 3, the predicted attenuation signals are in very good compliance with the measured values.

	tap water	salt water	bromo-benzene	edible oil
sound velocity c / ms^{-1}	1483	1780	1170	1440
relaxation magnitude A / dBs^{2}m^{-1} (Eqn. 6)	/	/	6.45E-12	6.20E-12
absorption coefficient B / dBs^{2}m^{-1} (Eqn. 6)	2.17e-13[1]	2.17e-13[2]	1.26E-12	3.60E-12
relaxation frequency f_m / Hz (Eqn. 6)	/	/	1.96E5	7.89E6

Table 1: absorption properties at ambient temperature (measured)

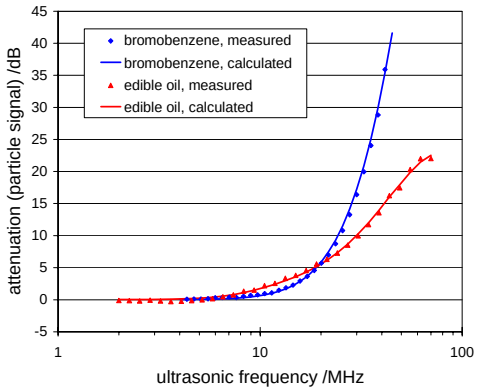

Figure 3: comparison of measured and predicted attenuation spectra

4.2 Detecting Trends in PSD, Comparison with Laser Diffraction

Using the experimental setup shown in figure 2, an emulsion of edible oil in salt water was produced. The pump rate was not modified over time.

Figure 4 presents the USE results of this measurement campaign in terms of the characteristic values x_{10}, x_{50} and x_{90} over time. As expected, the diagram shows a trend from coarse to fine particles. The shape of the characteristic values suggests to be asymptotic. About 30 minutes after starting the pump, measurements were stopped for adding tensides. Restarting the measurements, a further reduction of the particle size is obvious.

For comparing the USE results with LD measurements, three distinctive points were picked out. Within figure 4, they are marked out as ①, ② and ③. The corresponding cumulative distributions of the USE and LD measurements are shown in figure 5. It becomes apparent that both measurement principles indicate the trend in PSD nearly identically. The slight differences may be due to dilution effects during the LD measurements.

[1] literature value [6]
[2] measurements yielded no appreciable difference in comparison with tap water

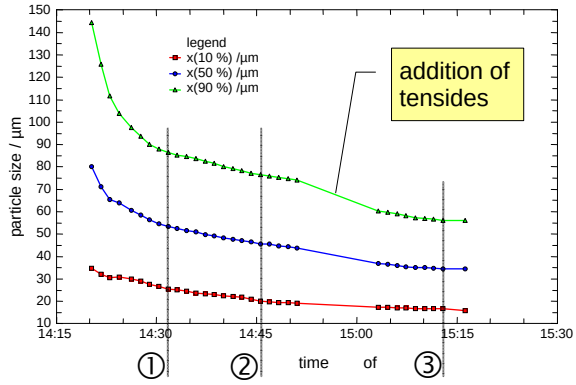

Figure 4: trend diagram of USE measurement results

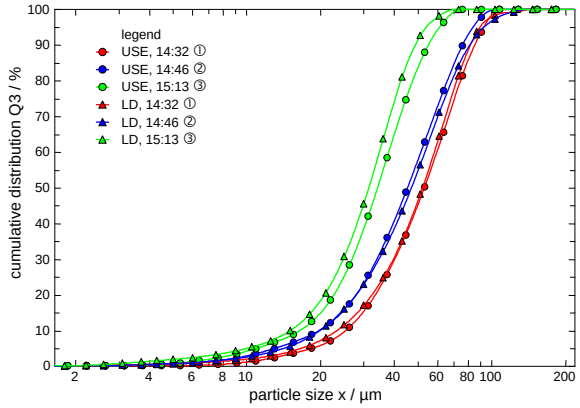

Figure 5: comparison of USE and LD particle size distributions

4 CONCLUSION

The presented results prove the worked out algorithm to be applicable for predicting the material dependent attenuation properties of emulsified systems. The calculated particle size distributions of ultrasonic extinction measurements meet all expectations regarding the comparability with laser diffraction results.

First industrial applications of the Sympatec OPUS System using the new algorithm are very encouraging.

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