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Measurement of the pure liquid's sound velocity in a bubbly sample volume

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Abstract:
This contribution will show how simultaneous group and phase velocity measurements can be used to calculate the sound velocity of the pure liquid, even if gas bubbles are interspersed. Apart from that it will be shown that for a given bubble size and operation frequency of an ultrasonic sensor only a certain volume fraction of gas is tolerable.

Key words: Sound Velocity, Bubbly Liquids, Concentration Measurement, Leaky Lamb Waves.

Motivation

The acoustic multisensor LiquidSens utilizes Leaky Lamb waves in a cavity for the measurement of temperature, density and sound velocity in liquids (Fig. 1). If the sensitivity of these variables to a change of predefined ingredients in a liquid mixture is high enough, LiquidSens can be used for a robust online concentration measurement. The robustness is due to the large aperture of the emitted Leaky waves in comparison to conventional focusing transducer setups. Therewith the sound wave has a good chance to reach the receiver, even if the liquid is interspersed with gas bubbles.

Fig. 1. Photograph of the acoustic waveguide sensor LiquidSens Probe.

Unfortunately the gas bubbles interact with the acoustic wave. They change the signals and therewith the measurable times-of-flight (TOF). This effect depends on the resonance frequency of the bubbles as well as the operating frequency of the acoustic sensor and can be misinterpreted as a change of concentration if not noticed. The aim of this contribution is to find unambiguous signal parameters for bubble indication or maybe a compensation of the gas bubbles disturbance.

Sound velocity in bubbly liquids

If two fluids are mixed together, e.g. water with a sound velocity of 1500 m/s and air with app. 340 m/s, the measurable sound velocity should be somewhere in between. But this is not always true, which has been shown in a long history of research starting in the late 1940's with Woods equation for free bubbles in a liquid. Therein, the sound velocity in a gas/liquid mixture is a function of bulk density and compressibility, implying that the operation frequency of the sonic measurement is far below bubble resonance frequencies and that there is no bubble interaction [1]. These conditions are mandatory to obtain the supposed intermediate sound velocities.

If the operation frequency is increased, the acoustic waves may excite bubbles. These can oscillate, store and release acoustic energy and thereby modify the measurable frequency dependent phase and group delays. This is why Lamarre and Melville measured higher sound velocities in bubbly water near the ocean surface. They explained these "sound-speed anomalies" with a phenomenologic dispersion model and with respect to their time-of-flight measurement [1].

With more understanding of the bubble-wave-interaction, that means thermodynamics [2], bubble dynamics, scattering and bubble interaction, Commander and Prosperetti developed a physical dispersion model for bubbly liquids. Additionally, they proved their model with some measurements, showing that the phase velocity in a bubbly liquid can be lower or higher than in a pure liquid [3]. The effect depends on the operation frequency of the sonic measurement and is highly associated with the measurable amplitudes. Finally it was to Orris et al. to include causality to the dispersion equation of Commander and Prosperetti [4]. Therewith, the interrelationship between phase, group velocity and attenuation holds in the entire frequency space. Apart from that the signal or energy velocity was introduced, to bridge the theoretical model to measurement setups.

For completeness it should be noted that there are further attempts to take also dispersed particles into account. For instance, Cents et al. have simultaneously measured bubble, drop and particle sizes in a multiphase system [5]. Therein, they also pointed out the resonance effect due to gas bubbles.

Linearized Orris-Model

The aim of this contribution is to transfer the theoretical model of bubble caused dispersion in a more simplified formula that is suited for integration in a measurement device. As Orris et al. have derived the most comprehensive dispersion equation we start here with the real valued part of their wavenumber $k(\omega)$ [4]:

$$k(\omega) = \frac{\omega}{c_0} \cdot \Re\left\{ \sqrt{1 + \frac{3\rho c_0^2 V}{\Phi - \frac{\rho a^2 \omega^2}{1 - j\omega \frac{a}{c_0}} - 4 j\omega\mu_0}} \right\} \tag{1}$$

With: $\Phi = f\left(\gamma_g, \kappa_g, p_\infty, T_g, \sigma, a, \omega\right)$

γ_g: Ratio of specific heats of the gas
κ_g: Thermal conductivity of the gas
p_∞: Ambient quiescent static pressure
T_g: Gas Temperature
σ: Surface tension
a: Unperturbed bubble radius
V: Volume fraction of gas
ω: Angular frequency
ρ: Liquid density
c_0: Quiescent sound speed
μ_0: Liquid‘s viscosity

LiquidSens operates at frequencies that are comparatively high to the bubble resonance frequencies. That means that the driving acoustic field is oscillating too fast for an efficient energy transport between the bubble and its surrounding fluid. Neither is there any viscous stretching of the liquid near the bubble‘s boundary [4]. This is why the Φ- and the μ_0-term can be neglected in eq. (1). The remaining high-frequency approximated wavenumber is dominated by radiation scattering and is only sensitive to the wavelength $\lambda_0 = 2\pi c_0/\omega$, the volume fraction of gas V and the bubble radius a:

$$\begin{aligned} k(\omega) &= \frac{\omega}{c_0} \cdot \Re\left\{ \sqrt{1 - \frac{3V c_0^2}{a^2\omega^2} \cdot \left(1 - j\, a \frac{\omega}{c_0}\right)} \right\} \\ &= \frac{\omega}{c_0} \cdot \Re\left\{ \sqrt{1 - \frac{3V}{4\pi^2 a^2} \lambda_0^2 \cdot \left(1 - j \frac{2\pi a}{\lambda_0}\right)} \right\} \\ &= \frac{\omega}{c_0} \cdot \Re\left\{ \sqrt{1 - \frac{3V}{a_n^2} \cdot \left(1 - j\, a_n\right)} \right\}, \quad a_n = 2\pi \frac{a}{\lambda_0} \end{aligned} \tag{2}$$

The complex root can be approximated if the radius a of the gas bubble is large in comparison to the wavelength, i.e. $a_n \gg 1$:

$$\begin{aligned} k(\omega) &= \frac{\omega}{c_0} \cdot \Re\left\{ \sqrt{1 - \frac{3V}{a_n^2} + j \frac{3V}{a_n}} \right\} \\ &= \frac{\omega}{c_0} \cdot \Re\left\{ \sqrt{1 - \frac{3V}{a_n^2}} \cdot \sqrt{1 + j\, a_n \frac{3V}{a_n^2 - 3V}} \right\} \\ &\approx \frac{\omega}{c_0} \cdot \sqrt{1 - \frac{3V}{a_n^2}} \cdot \left(1 + \frac{1}{8}\left(a_n \frac{3V}{a_n^2 - 3V}\right)^2\right) \end{aligned} \tag{3}$$

The phase velocity c_{ph} is the quotient of angular frequency and real valued wavenumber. The first-order Taylor series approximation for $V \approx 0$ (i.e. small volume fraction) leads to:

$$\begin{aligned} c_{ph}(\omega) &\approx c_0 \cdot \left(\sqrt{1 - \frac{3V}{a_n^2}} \cdot \left(1 + \frac{1}{8}\left(a_n \frac{3V}{a_n^2 - 3V}\right)^2\right) \right)^{-1} \\ &\approx c_0 \cdot \left(1 + \frac{3}{2}\frac{V}{a_n^2}\right) = c_0 \cdot \left(1 + \frac{3}{2} c_0^2 \frac{1}{\omega^2} \frac{V}{a^2}\right) \end{aligned} \tag{4}$$

The phase velocity in the high-frequency limit is always greater than the quiescent sound velocity. The deviation is linear proportional to the volume fraction of gas and reciprocal proportional to the cross section of a single bubble. Apart from that it should decrease with higher frequencies and lower sound velocities, i.e. shorter wavelengths of the acoustic wave.

The group velocity c_{gr} can be calculated in a similar way. It is the slope in the frequency-wavenumber plane:

$$\begin{aligned} c_{gr}(\omega) &= \frac{1}{k'(\omega)} \\ &\approx c_0 \cdot \left(1 - \frac{3}{2}\frac{V}{a_n^2}\right) = c_0 \cdot \left(1 - \frac{3}{2} c_0^2 \frac{1}{\omega^2} \frac{V}{a^2}\right) \end{aligned} \tag{5}$$

The absolute value of deviation is the same as for the phase velocity, but the sign is different.

We already know something similar from electromagnetics, where the geometric mean of group and phase velocity is the speed of light in vacuum. Therewith it should be possible to compensate the bubble dispersion by averaging group and phase velocity if the volume fraction is small enough and the measurement device is able to measure both, group and phase delay robust and precisely.

Parameter study

For the following numerical example the sound velocity is assumed to be $c_0 = 1482\,\mathrm{m/s}$. Fig. 2 shows the sound velocity deviations for two different bubble diameters, both, for the exact and the linearized model.

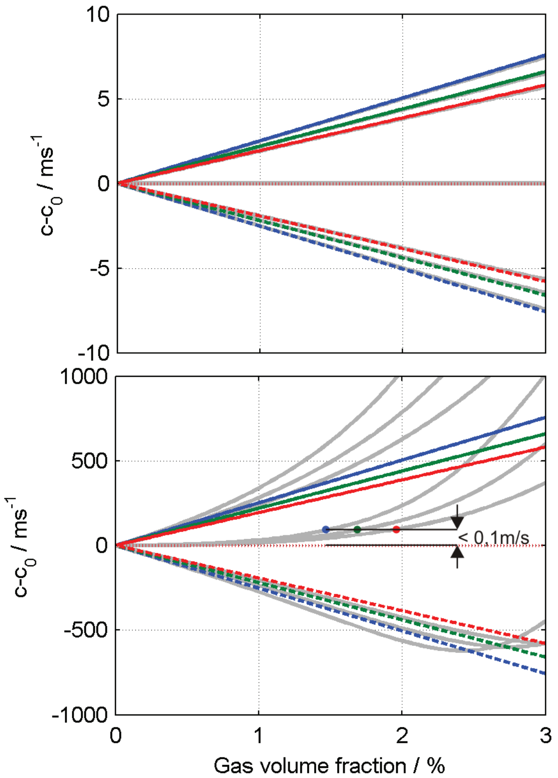

Fig. 2. Expected deviations of the measured sound velocities for gas bubbles with 1 mm diameter (top) and 0.1 mm (bottom); gray is the exact model value of eq. (1), colored are the results for the linearized model at different frequencies (blue: 1.4 MHz, green: 1.5 MHz, red: 1.6 MHz); solid lines are for the phase velocity, dashed for the group velocity – the averaged values are in between.

There is good agreement between the exact and the linearized model, especially for small gas volume fraction and large bubble diameters The idea now is to find the critical volume fractions V_{krit} that correspond to each bubble diameter in such a way that the absolute error of the averaged measured sound velocities, using the original model of eq. (1), is less than a certain limit, e.g. 0.1 m/s (see Fig. 2 and 3).

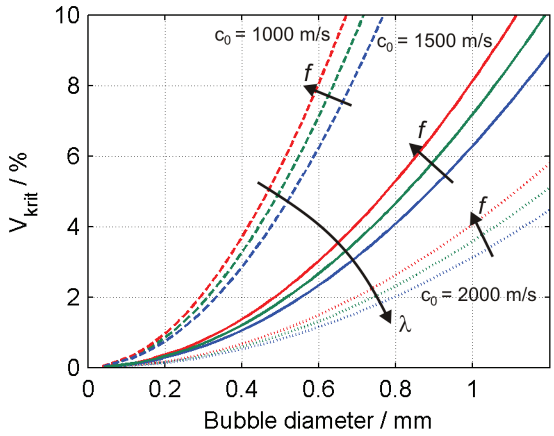

Fig. 3. Critical volume fractions for an accuracy of better than 0.1 m/s in sound velocity measurement as a function of bubble diameter for different sound velocities (dotted: 2000 m/s, solid: 1500 m/s, dashed: 1000 m/s) and different frequencies (blue: 1.4 MHz, green: 1.5 MHz, red: 1.6 MHz)

Fig. 3 demonstrates two important results:

a) With decreasing bubble diameter the maximal allowed gas volume fraction decreases.

b) The measurement gets more robust with shorter acoustic wavelengths.

Unfortunately the family of curves in Fig. 3 is hard to handle in a microcontroller. But if the normalized bubble radius a_{n} is used, the frequency dependence will be resolved. If furthermore the critical volume fraction is scaled with the square root of sound velocity, it is possible to parameterize a hyperbolic master curve:

$$V_{\mathrm{krit}}(a_{\mathrm{n}}) \cdot \sqrt{c_0} \approx \tilde{K} \cdot a_{\mathrm{n}}^2 \tag{6}$$

For the assumed maximum deviation of 0.1 m/s the parameter is $\tilde{K} \approx 0.283$. The parameterized curves of the volume fraction limits are in good agreement with the numerical results:

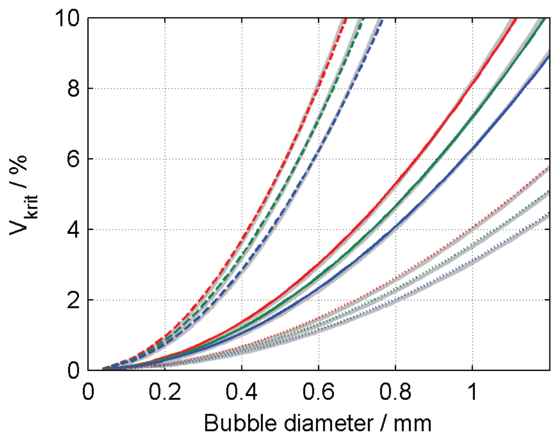

Fig. 4. Calculated (colored, same nomenclature as in Fig. 3) and parameterized critical volume fractions (gray), following eq. (6).

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Apart from the averaged sound velocity eq. (4) and eq. (5) can also be used to make an estimate about the bubble activity. The idea is to look at the relative span K between phase and group velocity:

$$K = \frac{c_{\mathrm{ph}} - c_{\mathrm{gr}}}{c_{\mathrm{ph}} + c_{\mathrm{gr}}} \approx \frac{3}{2} \cdot \frac{V}{a_{\mathrm{n}}^{2}} \qquad (7)$$

On the one hand the measured quantity K is a value for bubble activity at all. On the other hand, for precise sound velocity measurements it should always be less than a limit that can be expressed with the critical volume fraction of gas defined in eq. (6):

$$K = \frac{c_{\mathrm{ph}} - c_{\mathrm{gr}}}{c_{\mathrm{ph}} + c_{\mathrm{gr}}} < \frac{3}{2} \cdot \frac{V_{\mathrm{krit}}}{a_{\mathrm{n}}^{2}} \approx \frac{3}{2} \cdot \tilde{K} \cdot \frac{1}{\sqrt{c_0}} \qquad (8)$$

Fig. 5 shows three characteristic curves for the limits defined in eq. (8) and the expected values for a measured K, following eq. (1).

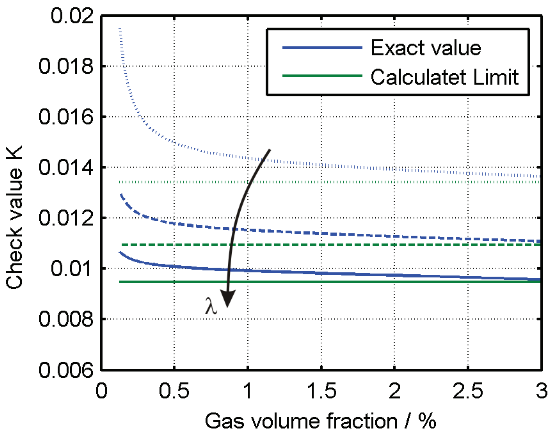

Fig. 5. Limits for too much bubble activity (green) and measurable parameter K for bubble check.

The calculated limit is always lower than the measured value of K. Therewith it should be a reliable boundary for too many bubbles in the liquid.

Laboratory setup and first results

For experimental verification a commercial available LiquidSens Probe Sensor was used. It consists of two parallel steel plates including the liquid under test. An initial ultrasonic Lamb wave propagates on one of these plates and partly leaks energy to the liquid. Therein the excited Leaky wave propagates zigzag between the plates and partly reconverts to Lamb waves with every nonspecular reflection. As a result the received signal consist of well separable signal packages on each plate (see Fig. 6) [6].

If bubbles are interspersed, only the first gray signal package in Fig. 6, correspondding to the first zigzag path, remains visible.

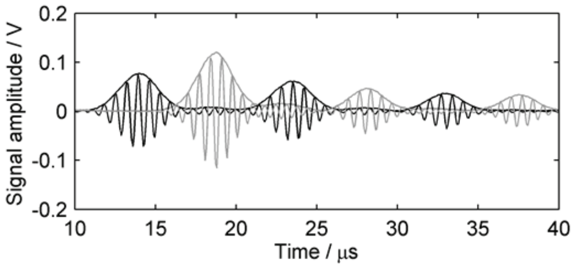

Fig. 6. Typical signals at the receiving IDTs for water at room temperature. Black: Plate with transmission IDT; Gray: On the opposite site.

Fig. 7 shows this signal package for different gas volume fractions in water and some photographs visualizing the bubble size and their distribution, approximately in a 1:1 scale.

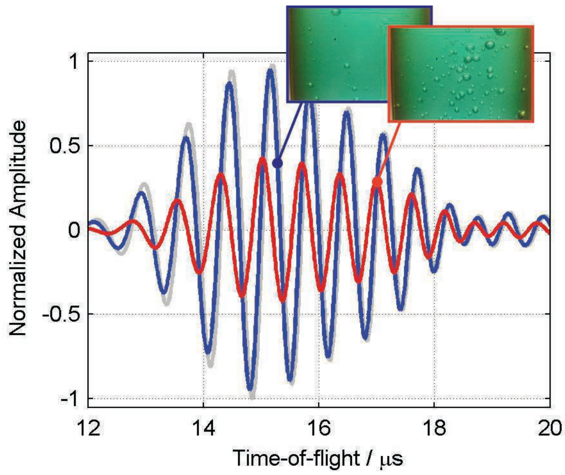

Fig. 7. The same first signal package without (gray) with little (blue) and high gas volume fraction (red).

The most obvious thing in Fig. 7 is the decaying amplitude if gas volume fraction is increased. Apart from that, the zero crossings shift to shorter times, which is evidence for an increased phase velocity. The envelopes are shifted towards higher arrival times (that means lower group velocities) and it seems as if the shift is just as big as the phase shift in the opposite direction (see Fig.8).

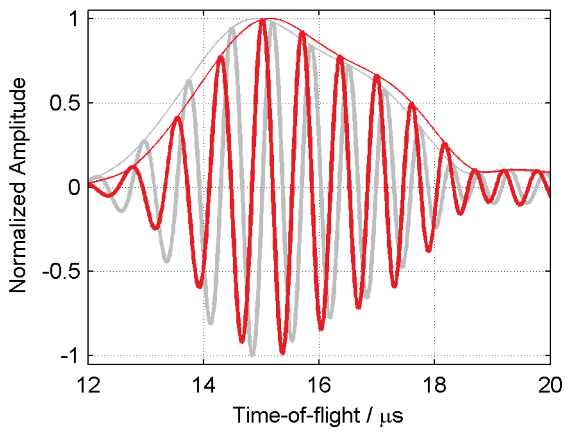

Fig. 8. The same normalized signal package without (gray) and with high gas volume fraction (red).

Offline TOF-Measurements

The first results have demonstrated qualitatively that the effect of eq. (4) and eq. (5) is measurable, but the robust simultaneous measurement of phase and group delay is still a challenge. On the one hand the group delay τ_g is defined as a frequency dependent function which has to be calculated as a phase-derivative in the frequency domain:

$$\tau_{gr}(\omega) = -\frac{d\phi(\omega)}{d\omega} \quad (9)$$

If the group delay should be approximated with only one value, it is necessary to modify the setup in such a way that the slope of the phase ϕ is almost constant in the frequency band of interest – we search for a linear phase system. On the other hand the phase delay is the ratio of phase and angular frequency:

$$\tau_{ph}(\omega) = -\frac{\phi(\omega)}{\omega} \quad (10)$$

If the phase is linear in frequency, as demanded before, and phase delay should also be constant in the considered frequency band, this implies $\phi(\omega = 0) = 0$. One possibility to ensure these signal properties is to use FIR-filters on the excitation signals. In such a defined system both, group and phase delay can be determined in the time domain using for instance quadrature amplitude modulated signals.

To check the measurement setup as well as the signal processing a two-step experiment was performed:

a) At time $t = 0$ gas is inserted in thermal oil with a small injector. Short time after gas insertion large bubbles float up with successive smaller inclusions.

b) After $t \approx 10$ minutes the gas valve is closed and the floating up of small bubbles decays.

Fig. 9 shows the corresponding time shifts with respect to their initial values. Both, group and phase delay show the expected characteristics. There is a sudden increase of the shifts directly after gas insertion and also the averaged shift immediately reaches another level which is nearly the same as after the degassing procedure. Then with a higher amount of smaller bubbles the shifts further increase and they start decreasing immediately after closing the valve.

Finally some remarks to the numerical quantity of the measured time shifts: For the liquid under test an error of only $0.05\,\mu s$ would result in a sound velocity error of more than 10 m/s. The bubbles influence in this experiment is approximately 10 times higher. Therewith a sole phase or group velocity measurement would fail in most applications.

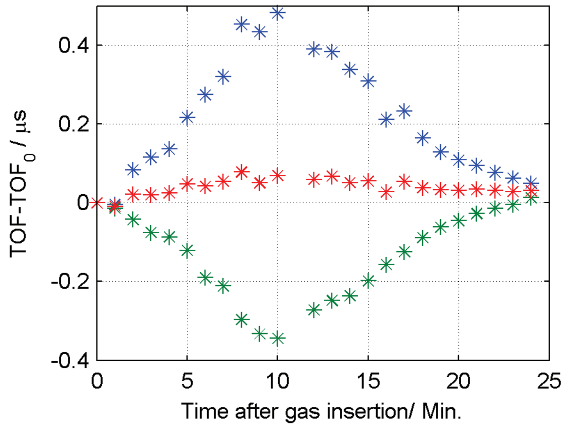

Fig. 9. Shift of group delay (blue) and phase delay (green) as well as the averaged shift (red) over time after gas bubble insertion.

Online Sound Velocity Measurements

As the algorithms for time-of-flight calculation were satisfactory they were implemented in a digital signal processor to provide sound velocity calculations with the proven LiquidSens platform. The prototypic setup was then put under test in a process where bubbles typically cause a standard system to fail because the sensitivity of sound velocity to a change of quiescent liquid properties is much smaller than the cross sensitivity to dispersed gas bubbles.

Fig. 10 shows the measured sound velocity of a standard LiquidSens sensor in comparison to the compensated sound velocity. Additionally the bubble indicator of eq. (8) was used to mask the invalid values. Due to the noisy process it was necessary to additionally reduce the factor for a satisfactory result.

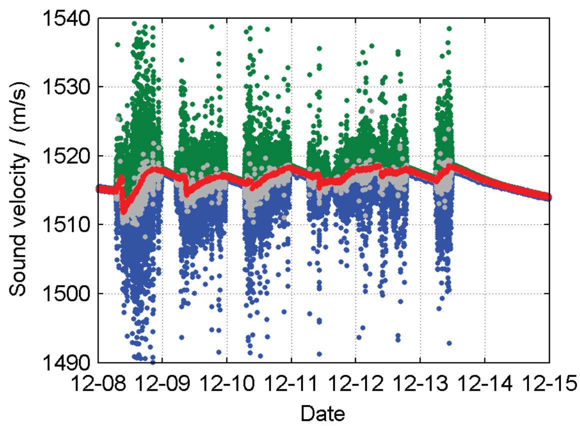

Fig. 10. Sound velocities in a bubbly process-water; green: standard LiquidSens phase velocity, blue: group velocity, red: pure liquid's sound velocity

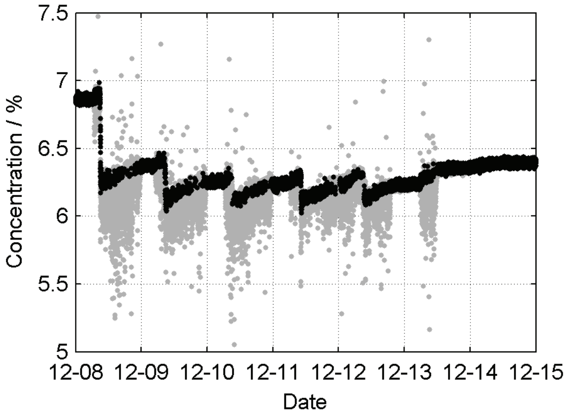

Fig. 11. Concentration values, calculated using temperature and averaged sound velocity of the prototypic LiquidSens Probe Sensor; black: valid sound velocities with little bubble activity; gray: invalid results

The averaged and valid sound velocities (see Fig. 10) can be used to calculate concentration values, whose variation is not larger than in the non-disturbed case (see Fig. 11). Therefor it was only necessary to adjust one factor $\tilde{K}$ (see eq. (8)) to match the process dependent bubble influence. Therewith the range of applications for ultrasonic process monitoring can be significantly enlarged.

Conclusion

Gas bubbles have high influence not only on the amplitudes but also on the times of flight in a liquid. The linearized Orris-model for the high frequency limit and small gas volume fraction shows that phase and group velocities drift with almost the same amplitude but different sign. Therewith it is possible to give a good approximation of the sound velocity in the pure liquid by averaging both measurements. The difference of phase and group velocity can be used to check for bubble activity in the liquid. Therefore, a boundary has been deduced that is reciprocal proportional to the root of the pure liquid's sound velocity and can be empirically matched to the process specific bubble activity.

Three measurements have proved the expected behavior. The first one has qualitatively shown that the phase arrival in a signal is earlier, whereas the signal envelope arrives later when bubbles are interspersed. The second one has demonstrated that simultaneous phase and group delay measurements in the time domain are suited to monitor the gassing and degassing in thermal oil and that the time shifts can lead to errors up to 100 m/s if sole phase or group delay measurements are used in bubbly liquids. The third, the online experiment has proved the applicability of the deduced and implemented formula. On the one hand the averaged sound velocity is a noisier but better value for the pure liquid sound velocity in a bubbly liquid. On the other hand the spreading of the measured sound velocities can be used to mask invalid measurement results.

The next step will be to average phase and group velocity in a more sophisticated way as both values have very different uncertainties. Maybe a Kalman-filter could be utilized for this purpose.

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