

ULTRASONIC CHARACTERISATION OF SOME AERATED FOODSTUFFS

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1 INTRODUCTION

1.1 Air in Foods

Many foodstuffs contain gas as a dispersed phase. Examples are ice-cream, meringue, bread, mousse, whipped cream and carbonated beverages. If the continuous phase is liquid, then the dispersion will be unstable. The principal destabilising mechanisms are

- creaming, due to the large density difference between gas and liquid,
- foam drainage (and subsequent collapse) due to the high interfacial tension,
- gas migration from the smallest bubbles into the larger bubbles due to the high Laplace pressure within small bubbles, via the continuous phase.

This last mechanism is termed *disproportionation*, and is an extreme form of Ostwald ripening. Clearly bubbly liquids are very dynamic systems, which makes the phase volume of incorporated air, ϕ_{air} , and the particle size distribution of the bubbles difficult to determine. Gravimetry is useful for determining ϕ_{air} , but it is insensitive and often hard to apply to foodstuffs. Microscopy is the method usually used to determine the bubble size distribution. The technique is hampered by the difficulty in representative sampling, and in the wide range of bubble sizes encountered in real systems [1]. This means that microscopy is at best a semi-quantitative method for characterising bubbly systems. There is a need for a technique which can characterise ϕ_{air} and bubble sizes in real time, and without the need for special preparation.

1.2 The Potential of Ultrasound

The usefulness of sound as a means of characterising bubbles has been recognised since the First World War, when the cavitation bubble clouds generated by submarine propellers were first used for submarine detection [2]. The difference in acoustic impedance between bubble and liquid leads to strong scattering, and the compressibility difference leads to a *giant monopole resonance*.

Minnaert [3] derived a simple expression to relate the resonant frequency to the bubble radius:

$$f_{\text{resonant}} = \frac{1}{2\pi r} \left(\frac{3\gamma P}{\rho} \right)^{0.5}$$

where r is the bubble radius, γ is the polytropic exponent of the gas in the bubble, P is the pressure, and ρ is the density of the liquid. A liquid containing a distribution of bubbles becomes highly dispersive and attenuating around the resonant frequency of the bubbles. There is an extensive literature on the theory of ultrasonic propagation through bubbly liquids, which has recently been reviewed by Commander and Prosperetti [4]. Figure 1 shows the predicted velocity and attenuation in water containing bubbles of $10\mu\text{m}$ radius at a ϕ_{air} of 10^{-2} . It can be seen that below the resonant frequency, the velocity in the bubbly system tends towards the simple volume average Wood velocity. Around resonance the velocity drops and then climbs to several times the value in the continuous phase. Above resonance the velocity

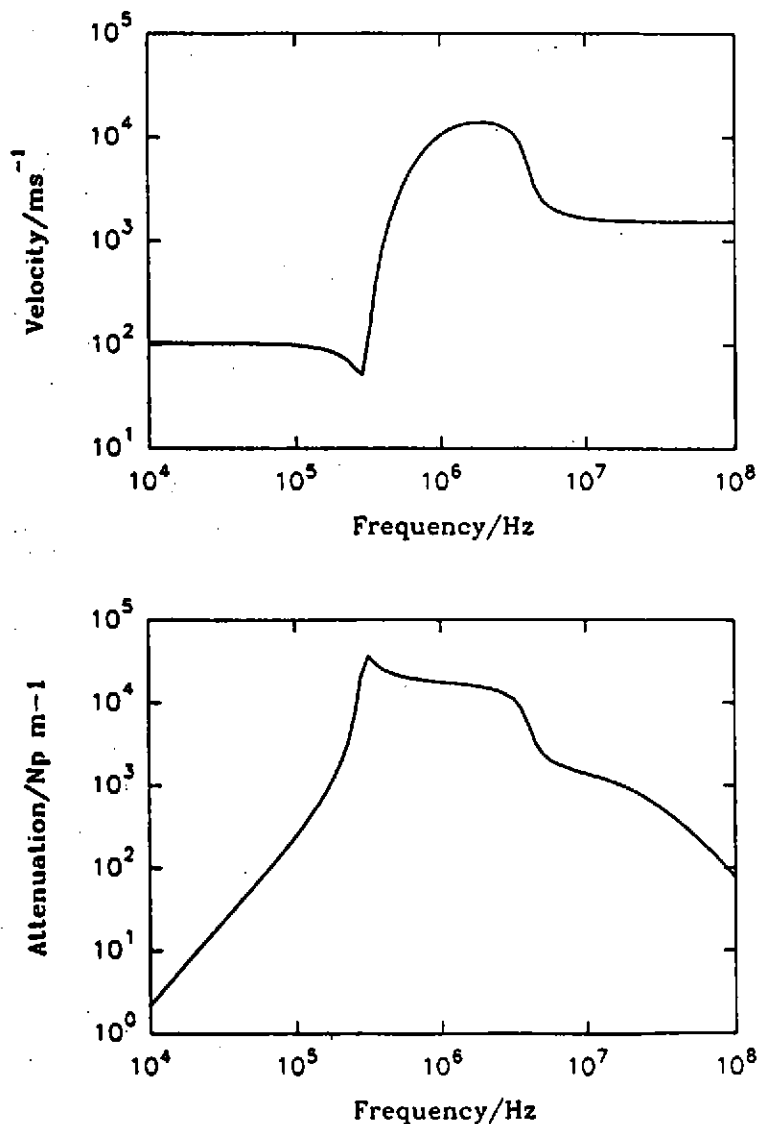


Figure 1. The top graph is the predicted velocity, and the bottom is the predicted attenuation in a monodisperse distribution of bubbles of radius $10 \mu\text{m}$ in water at a ϕ_{air} of 10^{-2}

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tends towards the value in air-free continuous phase. The attenuation climbs to very high values around resonance, reaching a peak at the resonant frequency. This demonstrates the sensitivity of ultrasound to bubble size, and hence the potential for bubble sizing.

1.3 Theoretical Descriptions

Figure 1 was prepared using the resonant scattering theory of Gaunaurd and Uberall [5]. This is a relatively simple effective medium theory. The derivation of the dispersion equation has been criticised [6],[7] but the theory has the advantage of computational simplicity. At low volume fraction the theory yields results which agree well with experiment. However, as the volume fraction increases, multiple scattering effects become more important and the theory becomes necessarily more complex. In order to calculate K , the propagation coefficient in the scattering medium, a knowledge of the single scattering coefficients, a_n , for an isolated bubble [8],[9],[10] is required. The a_n values are combined via a suitable radial distribution function [11],[12],[13] to yield a dispersion relation. This relation is analytic only for elastic scatterers in the long wavelength limit. For bubbles, numerical solution is necessary. The validity of the result depends critically on the form of the radial distribution function chosen, which places an upper limit on the ϕ_{air} values which can be handled. At very high volume fractions, the bubbles start to act as an ensemble [14], and a bubble cloud as a whole can have a series of resonances, which gives rise to additional scattering. The value of ϕ_{air} in aerated foods can easily exceed 0.5 and hence perfect agreement between results and measurements cannot be expected.

2 THE MEASUREMENT SYSTEM

2.1 The Measurement Cell

The measurement system is a development of a recently described instrument [18], and is shown in cross-section in Figure 2. The sample is held in an aluminium cell, which is thermostatted to $20.0 \pm 0.1^\circ\text{C}$. Two cylindrical perspex buffer rods are set into one wall of the cell, with waterproof transducers water-coupled to them (Karl Deutsch STS 40 WB, Sonatest SLH 5-20). Between them the transducers cover the frequency range 0.3-8 MHz. The measurement principle with each transducer is the same: A flaw detector (Balteau Sonatest UFD-1) is used to generate a 300V spike, which excites the transducer to produce a broad band pulse. The pulse travels down the buffer rod, and is reflected at the perspex-sample interface. On return to the transducer the pulse is detected, and analysed in the frequency domain via the Fast Fourier Transform (LeCroy 9420 oscilloscope), in terms of magnitude ($=r$), and phase ($=\theta$). The real part of the reflection coefficient of the sample is then given by $r \cdot \cos(\theta)$, and the imaginary part is given by $r \cdot \sin(\theta)$.

2.2 Interpretation of Measurements

The reflection coefficient, $R (=r \cdot \cos(\theta) + i \cdot r \cdot \sin(\theta))$ is related to the acoustic impedances (Z) of the perspex buffer rod and the sample by $R = (Z_{\text{perspex}} - Z_{\text{sample}}) / (Z_{\text{perspex}} + Z_{\text{sample}})$. Z_{perspex} was measured separately to be $3.245 \times 10^6 \text{ kg m}^{-2} \text{ s}^{-1}$ and R is measured and hence Z_{sample} can be calculated. If the value of ρ_{sample} is also known, then the relationships $Z = \omega \rho_{\text{sample}} / K$ and $K = \omega / c + i\alpha$ can be used to deduce the values of the velocity, c , and the attenuation, α , in the bubbly liquid.

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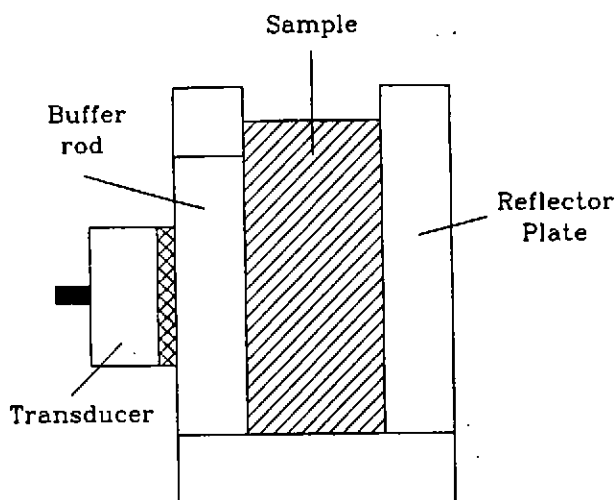


Figure 2. Cross-section of the measurement cell used in this work.

3 A MODEL BUBBLY FOOD SYSTEM

In order to facilitate comparison between measurement and theory, a simple model system was employed. The model system is required to be stable over the time period of the measurement (several hours), to be controllable, reproducible, and able to be characterised by non-ultrasonic means. The system chosen consisted of 'Sweetose' syrup (Ragus sugars, Slough, UK) at 70 wt% strength, whipped for one hour in a domestic Kenwood mixer, in the presence of surfactants. The surfactants were P1670 (Mitsubishi-Kasei Food Corporation, Japan) and ethanol, used at 1 wt% and 3 wt% respectively. P1670 is the sucrose ester of palmitic acid, and has an HLB number of 16. Sweetose was chosen because it is non-dispersive, very viscous (about 1000 times more viscous than water) and is not shear-thinning. The surfactant allows the formation of very small bubbles (by lowering the interfacial tension) and stabilises the bubbles against disproportionation by acting as a barrier to diffusion. The combination of viscous continuous phase and small bubble size results in a system with virtually no creaming. A particular advantage of using a large amount of surfactant is that it enables the bubbles to be sized in a Mastersizer. The Malvern Mastersizer MS20 measures particle sizes by means of laser light scattering. Because it works in the single scattering regime, it requires a great deal of dilution of the sample. Bubbles with radii of micrometre dimensions should dissolve very quickly in a liquid which is not supersaturated with gas. However, it is known that the presence of surfactant can stabilise even micrometre radius bubbles against solution [15]. The bubble size distribution was determined and found to give r_{32} values of around 10-20 μm . The size distribution data, along with a separate gravimetric measurement of the ϕ_{air} value (typically 0.5-0.55) and the appropriate thermophysical data were inserted into the theory due to Gaunard and Uberall [5]. Predicted velocity and attenuation are compared with a typical set of measurement data in figure 3. Reasonable agreement is obtained for the velocity figures, whilst the attenuation data shows relatively poor agreement. The results of these

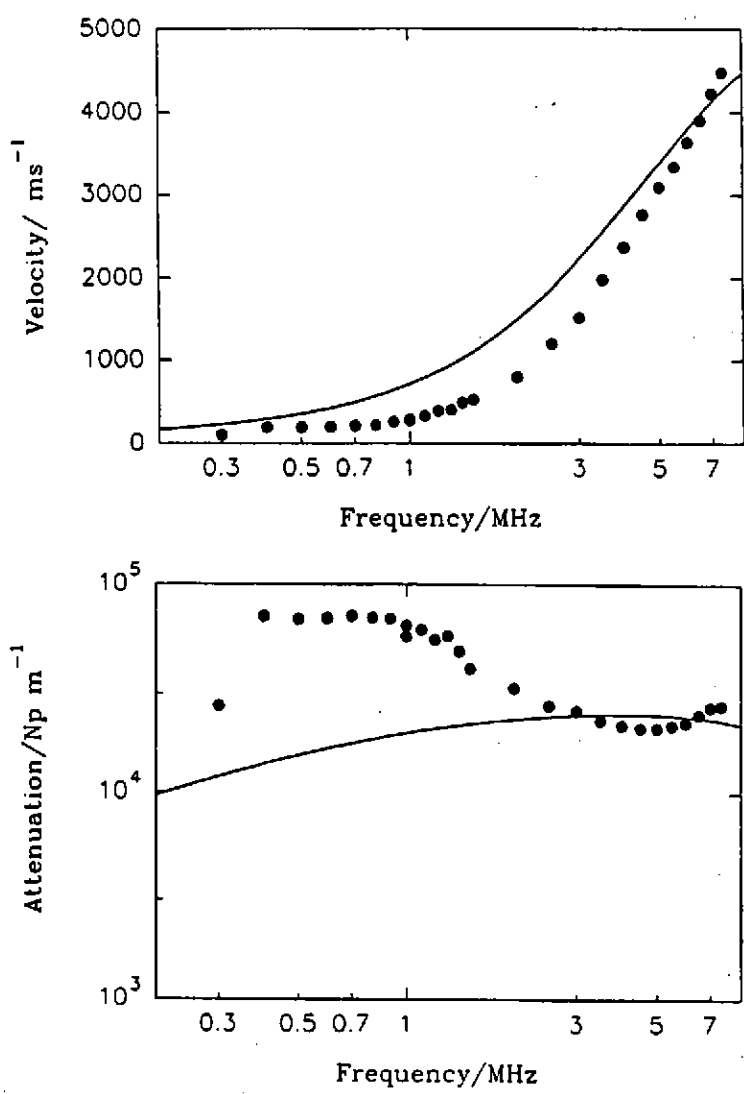


Figure 3. The top graph shows predicted velocity (solid line) and measured velocity (circles). The bottom graph shows predicted attenuation (solid line) and measured attenuation (circles).

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measurements are very encouraging for the prospect of characterising real foods by means of velocity measurements.

4 WHIPPED CREAM

Whipped cream is a complicated system, with two dispersed phases, one of which is semi-aggregated. In its unaerated form, whipping cream is a 35 wt% dispersion of fat in an aqueous serum containing lactose and protein. The fat is dispersed in the form of globules bounded by a lipoprotein membrane which protects the globules against coalescence, and helps render the smallest globules proof against even quite high shear. Whipping is a fairly mild, short time process which produces a wide range of bubble sizes. In the initial stages of whipping the bubbles are stabilized by protein, which is present in abundance. As whipping proceeds, the fat globules tend to move to the interface, and some of the larger fat globules are ruptured. The released fat promotes agglomeration of fat globules on the surface of neighbouring bubbles, causing a network to form which immobilises the bubbles and confers mechanical stability on the system [16]. If the temperature is too high, the free fat will be liquid, and will then displace protein and intact fat globules from the interface, thereby destabilising the air dispersion. Measurements were therefore made at $5.0 \pm 0.1^\circ\text{C}$. For increased stability, the cream was whipped past the point of maximum ϕ_{air} , at which point the viscosity increases markedly and the cream acquires a yield stress. Whipping conditions were 6 minutes at low speed in a Kenwood mixer at 4.5°C .

Measurements are presented in Figure 4 for the real and imaginary parts of the impedance of whipped cream (filled circles) and degassed unwhipped cream (hollow circles). It can be seen that the imaginary part of the impedance differs little between the whipped and unwhipped creams, and is quite large. This suggests that the unwhipped cream is quite highly attenuating, and that the bubbles in the whipped cream are relatively large ($r_{30} \approx 30\text{--}50\mu\text{m}$ [16]), such that the frequency range of the measurements is above the resonant scattering region. There is a clear difference in the values for the real part of the reflection coefficient. The velocity and density of the cream were measured separately at $\approx 1535\text{ ms}^{-1}$ and 1020 kgm^{-3} , giving a characteristic acoustic impedance of $\approx 1.57 \cdot 10^6\text{ kgm}^{-2}\text{s}^{-1}$. This compares reasonably well with the measured value of $\approx 1.67 \cdot 10^6\text{ kgm}^{-2}\text{s}^{-1}$. The ϕ_{air} was measured separately as 0.50. The measured value for the real part of the impedance of the whipped cream is difficult to interpret *via* theory, because of the polydispersity of the sample and the high volume fraction. If the velocity in the whipped cream is tending towards the value in the continuous cream, then the measured real impedance should be 0.50 times the value for the unwhipped cream. In fact the value is about 0.25 times this value. This could be because the assumption that the velocity returns to the continuous phase value is untrue, which has been suggested by Commander and Prosperetti. It has been shown [17] however that a good empirical relation can be derived between the real part of the impedance and the measured ϕ_{air} .

5 CONCLUSIONS

It has been shown that reflection measurements have the potential to provide useful information about aerated food systems, within limits. The limitations are the dynamic state of the sample, the difficulties in knowing the physico-chemical properties of the continuous phase to use in model calculations, and in making measurements by alternative methods to ultrasound in order to confirm the validity of the ultrasonically derived values. The difficulty in applying

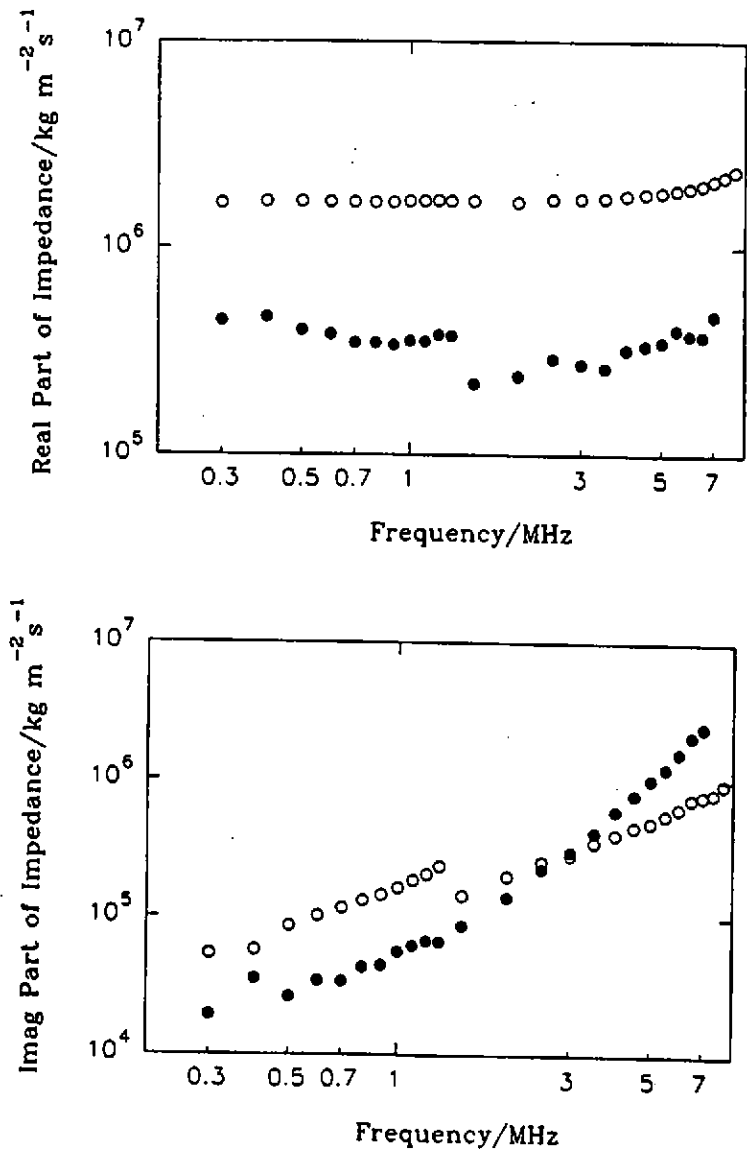


Figure 4. The top graph shows predicted velocity (solid line) and measured velocity (circles). The bottom graph shows predicted attenuation (solid line) and measured attenuation (circles).

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the theory is one of the biggest limiting factors in measuring foods. This work illustrates the difficulty in moving from a relatively well behaved model sample to a complex food stuff.

6 ACKNOWLEDGMENTS

We would like to acknowledge the financial support of the DTI, AFRC and Unilever plc

7 REFERENCES

- [1] R D BEE et al, 'Behaviour of an aerated food model' in Food Emulsions and Foams, p128, Ed. E DICKINSON, Royal Society of Chemistry, London, 1987
- [2] S M DOMENICO, 'Acoustic wave propagation in air-bubble curtains in water - Part I: History and Theory', *Geophysics*, **47**(3) p345 (1982)
- [3] M MINNAERT, 'On musical air bubbles and the sounds of running water', *London, Edinburgh and Dublin Philosophical magazine and Journal of Science*, **16** p235 (1933)
- [4] K W COMMANDER & A PROSPERETTI, 'Linear pressure waves in bubbly liquids: Comparison between theory and experiments', *J Acoust Soc Am*, **85**(2) p732 (1989)
- [5] G C GAUNAURD & M UBERALL, 'Resonance Theory of Bubbly Liquids', *J Acoust Soc Am*, **69**(2) p362 (1981)
- [6] L W ANSON & R C CHIVERS, 'Ultrasonic Propagation in suspensions - A comparison of a multiple scattering and effective medium approach', *J Acoust Soc Am*, **85** p535 (1989)
- [7] D J MCLEMENTS, P FAIRLEY & M J W POVEY, 'Comparison of effective medium and multiple-scattering theories of predicting the ultrasonic properties of dispersions', *J Acoust Soc Am*, **87**(5) p2244 (1990)
- [8] P S EPSTEIN & R R CARNHART, 'The absorption of sound in suspensions and emulsions. I. Water fog in air', *J Acoust Soc Am*, **25**(3) p 553 (1953)
- [9] J C F CHOW, 'Attenuation of Acoustic Waves in Dilute Emulsions and Suspensions', *J Acoust Soc Am*, **36** p2395 (1964)
- [10] R Y NISHI, 'The scattering and absorption of sound waves by a gas bubble in a viscous liquid', *Acustica*, **33** p65 (1975)
- [11] V TWERSKY, 'Acoustic bulk parameters in distributions of pair-correlated scatterers', *J Acoust Soc Am*, **64**(6) p 1710 (1978)
- [12] L TSANG, J A KONG & T A HASHABY, 'Multiple scattering of acoustic waves by random distribution of discrete spherical scatterers with the quasicrystalline and Percus Yevick approximation', *J Acoust Soc Am*, **71**(3) p552 (1982)
- [13] Y MA, V V VARADAN & V K VARADAN, 'Comment on ultrasonic propagation in suspensions', *J Acoust Soc Am*, **87**(6) p2779 (1990)
- [14] L D'AGOSTINO & C E BRENNEN, 'Acoustical absorption and scattering cross-sections of spherical bubble clouds', *J Acoust Soc Am*, **84**(6) p2126 (1988)
- [15] L A CRUM, 'Nucleation and stabilization of microbubbles in liquids', *App Sci Res*, **38** p101 (1982)
- [16] M ANDERSON & B E BROOKER, 'Dairy Foams' in *Advances in Food Emulsions and Foams* Eds E DICKINSON & G STAINSBY, Elsevier Applied Science, London (1988)
- [17] A MANETOU, 'Development of an ultrasonic sensor for monitoring air content', Leatherhead Food Research Association Report No. 667, Leatherhead F.R.A., UK (1990)
- [18] D J MCLEMENTS & P FAIRLEY, 'Ultrasonic Pulse Echo Reflectometer', *Ultrasonics*, **29** p58 (1991)