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Characteristics of Ultrasonic Scatter

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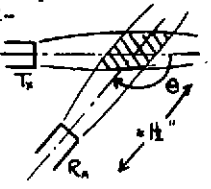
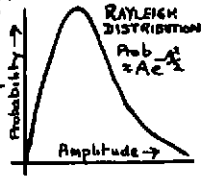
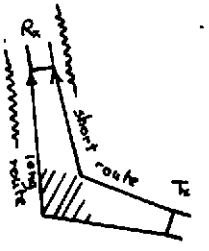
The purpose of this paper is to present some facts concerning the experimental observation of ultrasonic scatter from an emulsion; it is thought that this paper will be of general interest, as these observations are expected to be pointers in understanding other types of scattering system.

Hydrocarbon in water emulsions were made, stabilised by a soapy surfactant to prevent droplet coalescence. It is possible to vary the droplet size and the volume percentage of the hydrocarbon dispersed phase, and to show that the amount and type of ultrasonic scatter yields information about the structure of the mixture; this has obvious application to aspects of quality control of two phase media.

Two ultrasonic probes were immersed in the emulsion, on a probe holder that allowed one of them to rotate about the intersection region of their beam-patterns. This region is the "sensitive" volume, which can be examined at any desired "angle of scatter", θ. It is known (1, 2, 3) that the resultant amplitude of a sum of many small a.c. signals of random phase obeys a Rayleigh Probability Distribution law. If the transmitting probe were to send a continuous wave ultrasonic signal into the scattering medium, the receiving probe would have induced in it just this - a sum of many small a.c. signals, one c.w. signal from each scattering point; the phase would be random because this depends upon the transmitter-scatterer-receiver distance, and so a Rayleigh distribution should be obtained for the amplitude of the received signal. In practice, it is difficult to demonstrate this probability law with continuous wave ultrasound due to the difficulty of arranging that the receiving probe receives only those signals which are scattered from the sensitive volume. For the experiments reported here, a short burst of continuous wave ultrasound was used, the length of the burst being such that the latter end of the burst that went via the shortest possible transmitter - sensitive volume-receiver route overlapped with the front end of the burst that travelled via the longest possible route. In this way, the conditions of continuous wave working were simulated, without the disturbing feature of unwanted pick-up, sonic or electrostatic. The p.r.f. was 100 per second, low enough for stray reflections to dissipate before the next emitted burst. Harwell's submodular 2000 series N.D.T. electronics was used to give an output d.c. level proportional to the a.c. amplitude received when all parts of the sensitive



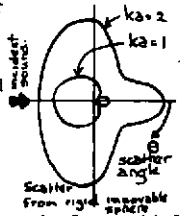




volume were contributing to the signal. For exploratory purposes, this statistically varying d.c. signal was analysed by a PDP8 computer, which calculated the mean value and plotted out the probability histogram obtained. In all cases the histogram was a good approximation to a Rayleigh Distribution, indicating that no stray signals were being received.

In a scattering measurement, the results must be randomised because the signal level varies statistically over wide limits, depending upon how constructively or destructively the many a.c. signals add. This can be achieved by moving the scattering medium relative to the probes or by sweeping the frequency over suitable limits. In the case of an emulsion, the randomisation takes place automatically by the former method as it swirls through the sensitive volume.

Scattering from a Single Particle. If a particle is small in comparison with the wavelength, the total scattered power is proportional to the square of its volume, and the polar scattering diagram varies in size but keeps its shape constant. When the particle size is comparable with the wavelength, the $(\text{volume})^2$ law breaks down, and the shape of the polar scattering diagram changes, developing side lobes. The characteristic shape is dependent upon the factor $\frac{2\pi a}{\lambda}$ (= ka), where λ is the wavelength and "a" the radius of the particle. The polar diagrams for the case of a rigid immovable sphere immersed in a liquid have been calculated (4); whilst these boundary conditions do not apply to the present case, something similar will happen.



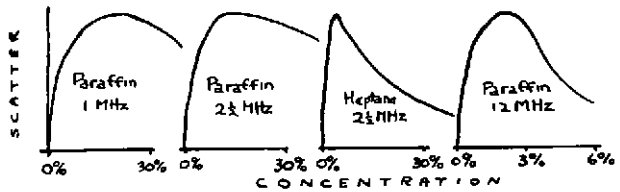
Scattering from many Particles. The scattering characteristics of a single particle are apparent in the way a suspension of such particles scatters. If a single particle scatters most sound into $\theta = 180^\circ$ (which is true if $ka < 1$), the mean scattered power of a suspension will be greatest at this angle also. The scattering polar diagram is changed in shape if there are particles in suspension for which $ka > 1$. The possibility arises of being able to determine particle size distributions through examining the shape and size of the polar scattering diagrams obtained from suspensions of particles.

The experimental observations on emulsions of liquid paraffin and of heptane in water were made in order to test the above hypotheses, and to high-light practical difficulties, and determine criteria by which the range of application of the technique may be evaluated.

Dependence upon Concentration of Scatters. From simple theory the relationship $(\text{mean amplitude}) \propto (\text{concentration})^{\frac{1}{2}}$ should be obeyed, being a consequence of the mean received power equalling the sum of the power scattered into the receiver by each scatterer in isolation. For the practical system of hydrocarbon droplets in water this relationship must break down, by the following considerations:-

1. At 100% concentration of hydrocarbon (no water), there is a homogeneous substance again, giving no scattering.
2. When the spacing between droplets is comparable with their size, their positions cannot be random, as each must fit into the interstices between the others; also many of them will be sitting in the "shaddow" of their neighbours.
3. Multiple scattering will take place, producing an apparent attenuation.

Amounts of emulsion were gradually added to distilled water in the scattering vessel until a 33% concentration by volume was obtained. Scatter was measured at $\theta = 140^\circ$. Both heptane and paraffin emulsions were used, each with a constant spread of droplet sizes in the ranges 0 - 150 μm. The frequencies used were 1 MHz, $2\frac{1}{2}$ MHz, and 12 MHz. The dependance of scatter amplitude upon concentration showed that the $(\text{concentration})^{\frac{1}{2}}$ law is accurately obeyed for small enough concentration. For increasing concentration, the signal increases less

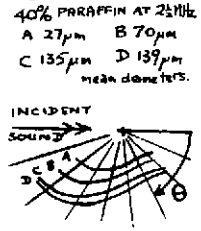


rapidly than a square root law, and a maximum amplitude is eventually reached. Beyond this, an increased concentration reduces the scatter signal. For paraffin, the concentrations giving the maximum signal were as follows:-

1 MHz	2½ MHz	12 MHz
20%	13%	2%

A plane water-heptane interface is a better reflector than a water paraffin, by a factor of 8½ times. It is not surprising to find, therefore, that a small percentage of heptane in suspension scatters a well as a larger percentage of paraffin. At 2½ MHz, and low percentage of heptane, the (concentration)$^{\frac{1}{2}}$ law is accurately obeyed and the maximum scatter amplitude is the same for both substances, but for heptane this occurs at only 1½% concentration by volume, as compared with 13% for paraffin.

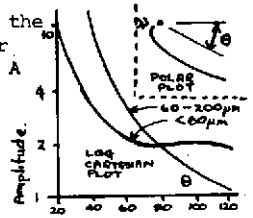
Dependence upon Mean Droplet Size. Forty per cent emulsions were prepared, of different average droplet size, and the polar scattering diagrams were obtained for these, at a frequency of 2½ MHz. Simple theory, which neglects attenuation effects, predicts that the power scattered from a suspension should be proportional to the mean droplet volume; consequently the volume weighted mean droplet size was used to characterise the suspensions, in order not to give undue emphasis to the many minute droplets that were always found. At 2½ MHz, practically all the scattering was Rayleigh "point" scattering, thus it was not expected that the polar scattering diagrams should differ in shape for the different emulsions. This was found to be borne out in practice, although the received power was less than linearly sensitive to the mean droplet volume (This would imply amplitude $\propto a^{3/2}$, whereas in practice it was found that amplitude $\propto a^{\frac{1}{2}}$ approximately). It must be remembered that at 40% suspension strength the full effects of a change in scattering ability are diminished through the attendant increased attenuation. At this concentration, there is a distinction in behaviour between changes in scattering ability due to concentration, and changes due to droplet size; increasing the concentration caused the received signal to diminish, whilst if the droplet size increases, the received signal increases.



Beyond the Rayleigh Region. As previously remarked, for Rayleigh scattering, the polar diagram shape is constant so it is not possible to know the "spread" in the droplet size distribution so long as all the droplets are small in comparison with the wavelength. However, if some of the droplets are larger than this (or the wavelength is reduced), the scattered energy shed from them will not go into the usual pattern obtained for Rayleigh scattering, and the existence of a fraction of larger droplets will be inferred by their polar diagram signature being impressed upon the total result.

To explore this hypothesis, the frequency was raised to 12 MHz (which makes ka = 2½ for 100 micron diameter droplets), and only emulsions with less than 2% paraffin were investigated, to minimise the unknown complication of the factors causing apparent attenuation. The polar diagrams for this frequency differed in character from the 2½ MHz; little energy was scattered in the backwards ($\theta = 180^{\circ}$) direction, the principal effect being to widen the beam, an effect previ-

ously noted by Allinson and Richardson (5) when the droplet size approached the wavelength, in their emulsion of a benzene - CCl_4 mixture in water. A preliminary experiment showed that the shape of the polar scattering diagram was independent of concentration, for a fixed droplet size distribution. Thereafter, two suspensions were made, one with droplets 60 - 200 microns in diameter, the other containing only droplets less than 60 microns. The concentration of both emulsions was adjusted to 1.9% and they were mixed together in a number of ratios to produce emulsions with different droplet size distributions. It was found that the "large-droplet" emulsion scattered more energy into the smaller angles of scatter ($< 80^{o}$) and less into the larger angles ($> 80^{o}$) than the "small droplet" emulsion. Composite emulsions, made from a mixture of both, demonstrated scatter characteristics intermediate between the two extremes and the ratio of the two components could be determined on the assumption that each contributed in proportion to its amount.



Summary. Theoretical predictions are well borne out at low enough concentration of dispersed phase, and an ultrasonic scattering technique may be made very sensitive to determinations of concentration or particle size. For higher concentrations (just how high depends upon the acoustic mis-match between the phases), a number of phenomena militate to reduce the received scatter signal; amongst these are the effect of multiple scattering which manifests itself as an increased attenuation, and the effect of the proximity of neighbouring particles upon the scattering contribution of any one of them. Although these effects cannot be isolated, it remains true that it is possible to monitor particle size even when the sensitivity of the technique to the concentration of the dispersed phase has vanished. Provided that particle size is small in comparison with the wavelength, the shape of the polar scattering diagram is invarient. For frequencies that give wavelengths such that $ka > 1$, the particles may no longer be considered as point scatterers; this has two effects a) viscosity losses cause an increased attenuation (5) and b) the shape of the polar scatter diagram is changed. However, for very dilute suspensions, the different scattering characteristics of differently sized particles can be used to obtain a measure of particle size distribution.

References

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