

Scattering of Ultrasonic Waves in a Liquid Mixture Near its Critical Solution Temperature

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INTRODUCTION

Certain mixtures of liquids exhibit the phenomenon of critical solution. At the critical solution temperature, two hitherto partially-miscible liquids mix completely; the meniscus between them disappears in a way similar to the effect occurring at the familiar liquid-vapour critical point. Ultrasonic propagation in such a mixture is usually studied by ordinary transmit-receive methods where attenuation is measured by the decrement of amplitude of an effectively non-divergent beam of ultrasonic energy. In such cases, a large increase of attenuation is observed in a small temperature range around the critical point (Chynoweth and Schneider, 1951).

It seemed possible, due to the inhomogeneity of the liquid mixture near the critical point, that scattering of sound might occur. This re-distribution of energy would appear as attenuation of the direct sound beam observed by the usual ultrasonic set-up.

EXPERIMENTAL

The apparatus was set up as shown in fig.1. The pulsed ultrasonic beam was radiated across the liquid-filled tank maintained at a constant temperature. The receiving transducer was carried on a cranked arm allowing a circular track to be traced out. The signal amplitude at angle θ measured from the axis of the direct sound beam was recorded.

The liquid mixture chosen was aniline-cyclohexane. This showed a critical point (above which complete miscibility occurred) at 30.5°C and 43% molar concentration of aniline. Several concentrations were examined at frequencies of 1.5, 2.5 and 5.0 MHz. Ultrasonic velocities lay in the range 1260 ms⁻¹ to 1420 ms⁻¹.

In an assumed non-scattering liquid such as benzene, having a sound velocity in the above range, the angular distribution of sound accorded well with the calculated diffraction pattern of the transmitter. For the mixtures, the distribution was much broader, indicating a scattering process (fig. 2).

ATTENUATION

The contribution of scattering to the overall attenuation in the liquid was calculated by estimating the total energy scattered at all angles after the direct sound beam had traversed a known distance. If this energy had been concentrated in the cone defining the major diffraction lobe of the beam, the direct ($\theta = 0^\circ$) acoustic signal would have been higher by a factor representing the scattering attenuation over the given distance. The energy estimates were performed by suitably weighting the angular distribution curves and integrating graphically.

These calculations showed that scattering was a major cause of attenuation quite near the critical point. Very close to that point, scattering appeared to be of smaller importance, an absorption mechanism coming into prominence (fig. 3).

SCATTERER SIZE

The experiments were done at frequencies 1.5, 2.5 and 5.0 MHz, so an investigation of the frequency dependence of the scattering attenuation coefficient (the "extinction coefficient") could be made (fig. 4). The similar problem of light scattering has been studied in great detail from the time of Rayleigh. Recourse to the literature revealed curves relating the scattering cross-section for light scatterers of varying refractive index to the parameter $2\pi a/\lambda$, a being the radius of a spherical scatterer and λ the wavelength of the radiation scattered, (Houghton and Chalker 1949) (fig. 5). The extension of this to the case of a sound wave was a formal matter, the frequency exponent in the "scattering law" shown by the slopes of the curves of fig. 4 being matched against the slope of an appropriate part of fig. 5. This allowed a value of $2\pi a/\lambda$ to be read off and a calculated. Some representative values are tabulated below for frequency 2.5 MHz and temperature $^{\circ}\text{C}$.

Molar %	$2\pi a/\lambda$	λ	a
Aniline	Estimated	(mm)	(μm)
22.5	1.5	0.50	120
33.3	2	0.51	150
43.8	3	0.53	250
53.9	5	0.54	400

Such a large size of scatterer seems at first a little strange. However, these conclusions are supported by investigating the spatial correlation of concentration fluctuations. This had been done for the optical case by Furth and Williams (1954) on the basis of the angular distribution of the scattered radiation.

The 'graininess' of the mixture has in fact been observed by Sliwinski (1964) using a Schlieren technique.

REFERENCES

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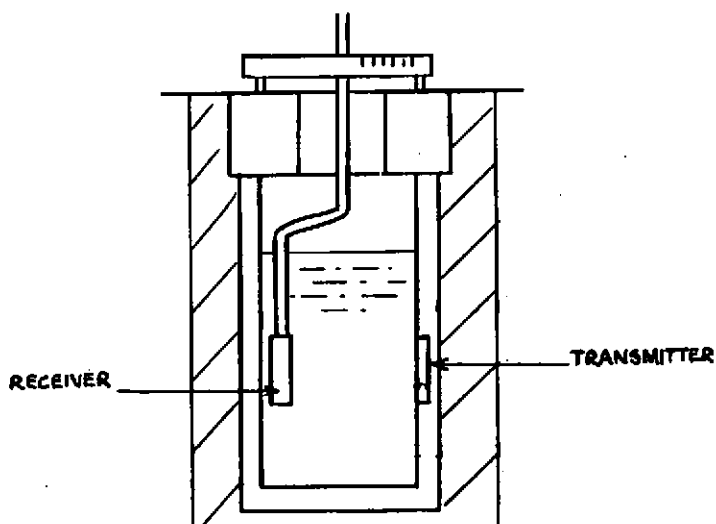


FIG. 1

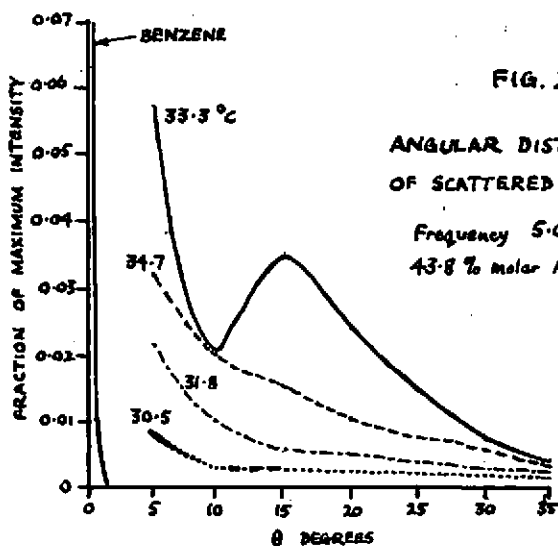


FIG. 2

ANGULAR DISTRIBUTION
OF SCATTERED SOUND

Frequency 5.0 MHz

43.8 % molar Aniline

