

THE PRODUCTION AND VELOCITY MEASUREMENTS OF ULTRASONICS IN SEVERAL LIQUIDS

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THE PRODUCTION AND VELOCITY MEASUREMENTS OF ULTRASONICS IN SEVERAL LIQUIDS

by

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The author would like to take this opportunity to recommend that anyone interested in the field of ultrasonics read <u>Ultrasonics and Their Scientific and Technical Applications</u> by Ludwig Bergmann. This book is a veritable gold mine of information to the research worker and shortcomings in theoretical developments are well made up for in the some five hundred and seventy-five references given in the bibliography. The author has made constant reference to this book **m** and instead of attempting to give credit in each place suggests here that if no other work be mentioned, Bergmann was consulted.

The author extends his sincere appreciation to Dr. J. W. McGrath who first aroused his interest in ultrasonics and who has guided this work. To list the others who have made suggestions, loaned apparatus and given encouragement would be simply to list the personnel of the department, and to all the author is grateful. A special word of thanks should be addressed to Prof. C. W. Chapman for his cooperation in the obtaining of apparatus and supplies.

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The Production and Velocity Measurements of Ultrasonics in Several Liquids

INTRODUCTION

Ultrasonic waves are, of course, sound waves of a frequency above the audible range. They are compressional waves, with frequencies ranging from 20,000 kcy/sec -just above the audible limit- up into the order of 10,000,000 kcy/sec. In air (v=331 m/sec) the wave-lengths then vary from the order of 1 cm to 10^{-4} cm. Thus the shorter ultrasonic waves are of the same order of magnitude as light waves. There is a rather great concentration of energy in these waves; Bergmann (1) shows that for an intensity of 10 watts/cm² the sound radiation pressure is 0.687 gr/cm². Intensities of this order are not at all unusual, as contrasted with audible sound in which intensities are on the order of 10^{-10} watts/cm².

By virtue of the short length of these waves, measurements which require awkward and unwieldy apparatus for audible sound can be carried forth in a much smaller space, as a number of wave-lengths can be produced where only a fraction of an audible wave could be present. Likewise with the short waves the influence of boundary walls is more easily removed. Laws valid for the audible range hold true for the ultrasonic range, although in the latter case phenomena appear which had not been observed in the former.

It is the purpose of this investigation to construct apparatus for the production of ultrasonic waves, and to make use of this apparatus to measure the velocity of sound in

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various liquids. The variation of sound velocity with temperature is of particular interest. It will be shown that the adiabatic compressibility and the ration of the specific heats of a liquid can then be calculated, which are otherwise rather difficult to measure and to compute.

THEORY

Richards (2) gives for a plane wave travelling in a homogeneous isotropic medium the equation

(1)
$$\frac{\partial \xi}{\partial t^2} / \frac{\partial \xi}{\partial x^2} = \frac{P_0 K}{P_0} \left(1 + \frac{\partial \xi}{\partial x} \right)^{-(\mu+1)}$$

in which f represents the displacement, x is the direction of propagation of the wave, P_0 and f are the pressure and density in the undisplaced portion of the substance, and f is the ratio of the specific heats. This equation is developed on the assumption that the sound wave is transmitted adaibatically. For very small amplitudes this equation gives the familiar result for the phase velocity

(2)
$$V_0^2 = \frac{P_0 t}{c_0} = \left(\frac{\partial P}{\partial c}\right)$$

in which $\frac{\partial P}{\partial \rho}$ is evaluated for the case $\rho = \rho_0$ The compressibility β is defined as

$$(3) \qquad \beta = -\frac{1}{\sqrt{2}} \frac{dv}{dp}$$

From this relation, it can be shown that the velocity of an elastic wave is also given by

(4)
$$V = \sqrt{\frac{V}{\rho \beta_{is}}} = \sqrt{\frac{1}{\rho \beta_{ad}}}$$

in which $\beta_{i,j}$ is the isothermal compressibility and $\beta_{i,j}$ the adiabatic compressibility.

It is this last relation which gives to sound velocity measurements one of their chief uses, for the adiabatic compressibility together with the isothermal compressibility allows us to calculate directly the ratio of the specific heats. From this, we can calculate the specific heat at constant volume, which otherwise involves complicated thermodynamical relations.

In 1932 Debye and Sears (3) and also Lucas and Biquard showed independently of one another that light passing throught a liquid traversed by a sound wave is diffracted and phenomena appear very similar to those obtained with an ordinary transmission grating. There are apparently two effects; the periodically changing index of refraction bends the waves, and also there is a periodic retardation of the light in various parts of the sound beam. The effective grating space is the distance between successive compressions, or the wave-length of the sound Λ . It is shown that the ordinary grating law holds, or

(5)
$$n\lambda = \Lambda \sin \theta$$

where n is the order of the diffraction image, Θ is the angle of diffraction for the nth order, and λ is the wave-length of

the light. Eq (5) combined with the expression for the velocity of a wave $v=\Lambda v$ gives for the velocity of sound

(6)
$$V = \frac{n\lambda v}{\sin \theta}$$

Obviously the deviation of the orders depends upon the frequency of the sound, higher frequencies causing greater deviation. For example, in liquids the velocity is on the order of 1200 m/sec, and for a frequency of 1800 kcy. the deviation is on the order of 1.5 mm. at a distance of 200 cm. This consideration shows that impossibility of using audible sound in this manner, for a frequency of 20 kcy. would give a deviation of about 0.0016 mm, and lower frequencies even less.

This method gives us an easy way to measure the velocity of sound, for the diffraction pattern may be preserved on a photographic plate and the deviation of the orders measured with considerable accuracy on a comparator. The other measurements required are the distance of the plate from the lens, the frequency of the sound (the frequency of the driving oscillator) and the wave-length of the light, all of which can be made to a fairly high degree of accuracy.

Either standing or travelling sound waves may be used in this method, the diffraction effects being affected only slightly. In the case of travelling waves the grating is moving across the light beam with the velocity of sound; in the case of standing waves the grating is oscillating back and forth across the light beam through a distance A_1 .

APPARATUS

There are several methods of producing ultrasonic waves, of which probably the most convenient for the majority of purposes is the piezo-electric effect of quartz crystals. It is well known that if a difference of potential be applied to opposite faces of a slab of quartz the slab will either expand or contract in the direction normal to those faces, depending upon the sign of the charge. If this differences of potential be alternating the slab will vibrate, the vibrations being maximum when the impressed frequency is equal to the natural mechanical frequency of the quartz. The faces of the slab thus give off sound waves, the frequency being determined by the thickness of the slab.

The accuric crystal used in this investigation was an ordinary one, produced for the control of oscillator circuits. An X-cut crystal, was used as this type is thick enough to be fairly rugged. The dimensions were 2.5 x 2.5 X .11 cm. The temperature coefficient was given as -15 cy/meg./deg. C. A layer of gold was evaporated on both sides of the crystal to provide better electrical contact, and the high frequency voltage was supplied to the crystal through the holder shown in Fig. I. The crystal was held against a brass plate forming one of the electrodes by a light phosphor bronze spring, which acted as the other electrode. The end of the spring was divided as shown and contact was made at about the vertical center of the crystal in order to avoid as far as possible distortion of the sound field due to uneven mechanical damping of the crystal. The whole thing was then mounted on a bakelite backing and clipped to one end of the trough which held the liquid.

The trough is shown in Fig. II. It is simply a brass box with plate glass sides to avoid optical defects present in ordinary glass. It is important that the sides of the glass plates be parallel, as well as the plates themselves in order that optical effects further then those due to the sound beam be eliminated. The dimensions of the trough are relatively unimportant, except that it should be somewhat larger in crosssection than the sound beam so that the spreading of the beam is not affected by the boundaries.

As the far end of the trough was placed an acœustic absorber, modelled after one described by Fox and Rock(4), and shown in Fig. III. It is a brass cylinder with an opening in the side, around which is built a collector to direct the sound beam inside. A brass reflector is placed inside and is skewed with respect to the opening, so that the sound is reflected up and to one side, the energy being dissipated by multiple reflections within the cylinder. This absorber was used to minimize the effect of standing waves due to reflections at the far end of the trough, which would tend to confuse the sound field.

Stirring the liquid should not affect the sound field, and would remove some of the effects of large temperature gradients due to high intensities between the sound beam and the rest of the liquid as well as within the beam itself. To this end a mechanical stirrer was made, consisting of a propellor blade driven by a motor which was rated at 1600 rpm.

This motor was obtained through the courtesy of Mr. G. W. Hoddy of the A. G. Redmond Company, Owosso, Michigan. However, it was found that the flow of liquid was neither rapid enough nor smooth enough to completely remove all thermally produced density imhomogeneities, and the result was a blurring of the pattern. In order to avoid this effect and yet to have better thermal conditions the liquid was stirred immediately before a photograph was taken, the temperature recorded, stirred again after photographing, and the second temperature recorded.

Temperatures were measured with a thermometer graduated in tenths of a degree Centigrade, which was found to read about 0.3° low at the boiling point of water, but was all right at 0° C. The thermometer was placed centrally but not in the beam.

The optical system was like that used for an ordinary grating. The source of light was a high pressure mercury arc, filtered so as to cut out the blue, most of the red and part of the yellow. It was found that if the arc was filtered so as to eliminate all but the green not enough intensity remained to photograph in any reasonable length of time, The light was focussed on an adjustable slit by a condensing lens, and rendered parallel by an achromatic combination. After passing through the trough and being diffracted, the light was brought to a focus on the photographic plate by means of a +200 cm focal length spectacle lens. The optical set-up is shown diagrammatically in Fig. IV.





The acoustic crystal was driven by a crystal-controlled power oscillator, whose circuit is shown in Fig V. The oscillator tube was an RK-20A, a pentode, and was operated with a d.c. potential of 1000 volts on the plate, +300v on the screen grid, and +45v on the suppressor grid. The feedback condenser shown connected between the plate and the control grid was simply two insulated wires twisted together, as it was found that any more capacitance gave too much feedback.

The control crystal was rated at 1806 kcy $\pm 0.03\%$. The manufacturers recommended that 150 milliamperes by the maximum current across the crystal, and to this end a 125 ma. fuse was installed in series with the crystal. Many helpful suggestions as to the construction of this oscillator were obtained from The Radio Amateur's Handbook (5).

Radio frequency power was transmitted from the oscillator to the acoustic crystal circuit by means of a link coupling, also shown in Fig. V. The links at either end consisted of three turns of #14 tinned copper wire, and the transmission line was simply a length of ordinary lamp cord. This method allows of transmission at low potentials, mobility of the acoustic crystal unit, and still permits relatively close coupling. The links were mounted inside the two tank coils in such a manner that they could be turned in order to vary the coupling if necessary. In this case, as maximum power pransfer was not particularly desired, no attempt was made to obtain critical coupling. The maximum coupling possible was used at all times, and even then the coupling was relatively loose.

The acoultic crystal circuit shown in Fig. V reproduced as exactly as possible the tank circuit of the oscillator, in order that the impedances be equal. The crystal was then shunted across the circuit. In both tank coils the coupling links were mounted on the ground end of the coil, so that the transmission line was not at a high potential with respect to the ground.

The power supply for the oscillator consisted of a fullwave rectifier circuit utilizing two 866 mercury vapor diodes, as shown in Fig. VI. The choke-input filter circuit shown is calculated to give a ripple of approximately 0.75%. The 25,000 ohm bleeder resistance in the power supply serves three purposes: it supplies a minimum load, allows the filter condensers to discharge without arcing when the load is removed, and also acts as a potential divider to supply the various voltages for the RK-20A.





FIG. VI POWER SUPPLY CIRCUIT

EXPERIMENTAL PROCEDURE:

With this frequency of 1806 kcy/sec the angle of deviation of the diffraction orders is so small that it was found necessary to use a very long focal length objective lens. The lens used was a spectacle lens of focal length +200 cm, which gave a deviation on the order of 1.6 mm. The collimating lens was an achromatic combination of focal length 20 cm, and a very small difference in the position of this lens made a large difference in the position of the image. To avoid this error the plate holder was set 200 cm from the objective lens, and the collimator adjusted until the image was in focus. This could be done as the focal length of the objective was known rather accurately, and for a focal length of this magnitude, a slight error in the setting of the objective affects the position of the collimator very little. Adjustment in this manner appeared to be the easiest and most accurate way of insuring that the light incident upon the trough be parallel. Care was taken that all the components of the optical system be at the same height, and perpendicular to the path of the light.

It was shown by Brillouin that the intensity of the diffraction orders depends upon the intensity of the sound wave (see Bergmann, pp. 72-3). This, in turn, depends upon the power supplied, the distance of the light beam from the crystal, and upon the liquid in which the sound field is set up.

The closer the light beam is to the acoustic crystal the more intense are the higher orders, but also the sound field is somewhat distorted in the immediate neighborhood of the source. This last is due to the fact that the crystal does not vibrate exactly like a piston even under good conditions as well as to the fact that the effective acoustic aperture is large compared to the distance away. In this work the light beam passed through the liquid five centimeters in front of the crystal, in an attempt to compromise between these two effects.

Eastman Spectroscopic plates V-D were used, and exposure times were on the order of five minutes in most cases. Some photographs were obtained in times as short as forty seconds, however. Exposure times were made as short as possible in order to avoid large temperature differences during the exposure.

The tank circuit of the oscillator was tuned slightly off the maximum dip in plate current, as at the critical point the oscillator is somewhat unstable and the current through the control crystal is greater. The same procedure was used in the acoustic crystal circuit, although here the indication of resonance was a rise in the crystal current. For most of the measurements the r.f. anmeter indicated about 0.3 amperes, although it is questionable as to whether this was actually the current through the crystal. There was a relatively large amount of energy radiated from all parts of this circuit - the tank coil, leads, and even the trough in which the sound was generated.

The combination radio frequency standard and mixer -

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amplifier circuit was used to calibrate a continuous range oscillator which in turn was used to measure the frequency of the voltage supplied the acoustic crystal. Signals from the calibrated oscillator were mixed with those from the test oscillator in the combination instrument and tuned to zero beat. Several measurements made in this manner gave 1806[±] 4 kcy/sec as the frequency of the acoustic system. No difference in frequency was detectable when the sound field was turned on or off.

In order to provide a uniform distribution of charge over the quartz crystal, used as the source of sound, a coating of gold was evaporated on each face. 24k gold was obtained from a dental supply house in the form of small pellets, and only small amounts were used to give an adequate coating. The actual evaporating was done in a vacuum system, and strips of Scotch tape were used to prevent the gold depositing around the edges and so forming a short circuit when in use.

The distance d in Eq. (10) were measured on Gaertner comparator which was graduated to read direct to thousandths of a millimeter. With this particular instrument all readings had to be taken from low to high numbers and the platform run in only one direction to eliminate the effect of backlash in the screw. In most of the plates the central image was overexposed so the distance between right and left orders was measured and half this distance taken as the deviation d. The edge of the orders were in most cases not well defined so measurements were taken from the centers. To compensate for random setting on the centers ten readings were taken for each of two positions on the plate and the mean of the twenty read-

ings used as the value of d. For each liquid two plates were taken at as nearly the same temperatures as possible in order to provide a check. In the case of propyl alcohol at a temperature below room temperature only one plate was taken as it was found that moisture from the air condensed on the sides of the trough and rendered the image too diffuse for photographing.

The acoustic power input into the trough was measured as suggested by Bergmann (p. 37). The liquid, in this case butyl alcohol, was heated by an electric heater through about the same temperature range in the same length of time as was caused by the sound and the electric power measured. This method assumes that most of the sound energy is converted into heat in the liquid, which is most probably the case. The index of refraction of the liquid, the brass sidewall and the air are all quite different, and it can be shown that the amount of reflection depends upon the difference in the indices of refraction of the media. However, much of the energy from the oscillator is lost before it is converted to sound, the electromagnetic radiation in the neighborhood of the crystal leads and the brass trough being sufficient to light a neon test bulb. Thus although the RK-20A tube is rated at 50 watts output, measurements of the acoustic power indicated only about 7.7 watts.

Velocity measurements were made on butyl alcohol, and vinyl acetate at three different temperatures - one at room temperature, one below room temperature, and one several degrees above room temperature. As far as could be determined

no measurements had been made on vinyl acetate, and information as to the temperature dependence of sound velocity in the others was unavailble. Measurements were also made on ethyl alcohol at room temperature.

DATA AND RESULTS

Ethyl Alcohol C2H50H

Plate No.	18	<u>19</u>
t ^o C	23.0	23.5
d (mm)	1.549	1.562
人 (cm)	0.0705	0.0698
v (m/sec)	1272	1261
dens. (g/cc)	0.7869	0.7865
β_{ad} x 10 ⁶ atm ⁻¹	79.61	82.55
βis x 10 ⁶ atm ⁻¹	100.68	101.09
'r	1.26	1.23

n-Propyl Alcohol C₃H70H

Plate No.	72	-1	વ્ય	42	43
Mean t ^o C	13.2	30.8	30.5	40.7	40.5
d (mm)	1.575	1.635	1.637	3.360	3.350
Å t (cm)	0.06938	0.0668	0.0667	0.0650	0.0652
(uss/m) N-	1252	1206	1205	1174	1177
dens. (g/cc)	0.8095	0.7956	0.7958	0.7877	0.7878
$\beta_{ad} \ge 10^{6} a tm^{-1}$	79.82	87.54	87.54	93.29	92,88
$\beta_{1s} \ge 10^{6} \mathrm{atm}^{-1}$	X102.3	104.2	104.1	105.3	105.24
2	1.29	1.19	1.19	1.13	1.14

) ₃ 0H
CH2
CH ₃ (
Alcohol
n-Butyl

Plate No.	26	27	ဗ၊	41	<u>40</u>	4
Mean t ^o C	12.0	14.1	26.9	27.3	41.1	40.3
d (mm)	1.403	1.4 35	1.582	1.583	3.201	3.197
A (cm)	0.0778	0.0761	0•0690	0 • 0690	0.0682	0.0683
v (M/sec)	1405	1374	1248	1246	1232	1233
dens. (g/cc)	0.8153	0.8140	0.8053	0.8042	0.7935	0.7941
/3 ad x 10 ⁶ atm ⁻¹	62 .91	65.90	81.23	81.16	84.13	83,83
$\beta_{1s} \times 10^{6} \mathrm{atm}^{-1}$	87.51	89.15				
<u>~</u>	1.39	1.35				

Vinyl Acetate

Plate No.	45	46	<u>81</u>	38	37	39
Mean toC	19.9	18.8	24.1	25 . 4	39 ° S	38.1
đ (mm)	3 .396	1.670	1.763	1.74 6	1.847	1,840
🔨 (ст.)	0.0643	0.0654	0.0613	0.0625	0 .0591	0.0593
v (m/sec)	1161	1181	1106	1129	1067	1072

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DISCUSSION OF RESULTS

Values listed for densities were obtained through the use of extrapolation formulae given in the International Critical Tables (6). Values for the isothermal compressibilities were also obtained from this source.

Isothermal compressibilities for butyl alcohol were given only for 3.05° C and for 17.4° C. These values were interpolated to 12.0° and 14.1° C and l' calculated for these temperatures. In the absence of more reliable temperature variations relations, l' was not computed for the other plates. Density and compressibility values for vinyl acetate was not listed, so that no calculations other than velocities could be made.

The adiabatic compressibilities were calculated from Eq. (4). This equation shows that the ratio of the specific heats is also given by the ratio of the isothermal to the adiabatic compressibility. As the usual way of expressing compressibilities is in terms of reciprocal atmospheres, the values given by Eq. (4) must be multiplied by 1,013,000 (76 x 13.60 x 980).

There are three measurements of importance in the calculation of velocities: first, the frequency was measured as described towithin about 0.25%; second, the distance from the objective lens to the photographic plate could certainly be measured to within 0.5 cm or 0.25%; third, the distance d on the plates could be measured to within 0.008 mm or 0.5%. This gives an estimated maximum error of one percent. However, results did not check this closely with those given in Bergmann (p. 122-3-4-5). Bergmann gives 1234 m/sec for

propyl alcohol at 24°C, while interpolation from results obtained here gives 1223 m/sec, or a difference of about 0.9%. For butyl alcohol at 23°C, Bergmann gives 1315 m/sec, as compared to 1272 m/sec obtained here. This a difference of about 3.3%. One measurement on ethyl alcohol gives 1261 m/sec at 23.5°C, while Bergmann gives 1207 m/sec, for a difference of 4.5%.

This estimated error will be just doubled for the adiabatic compressibilities and the ratio of the specific heats, for the velocity enters here as the square. For ethyl alcohol at 23.5°C Bergmann gives for / 1.214, and this determination gives 1.23. Bor butyl alcohol at 23°C Bergmann lists / as 1.160, and these results give 1.35 at 1.41°C. For propyl alcohol Bergmann gives 1.162 at 24°C, and this investigation gives 1.19 at 30.8°C. Rough interpolation to 24°C gives 1.23, or about 7% difference.

 β_{ad} for propyl alcohol at 24°C is given by Bergmann as 82.9 x 10⁻⁶ atm⁻¹, and results here give 84.8 x 10⁻⁶ atm⁻¹ or 2.5% difference. β_{ad} for butyl alcohol at 23°C is given by Bergmann as 72.5, while these results give 78.0 for a difference of about 6.2%. For ethyl alcohol β_{ad} is listed as 88.3 at 23. 5° C, as compared to 82.55 obtained here, or a difference of 6.5%.

An explanation of these differences which are larger than expected is not readily apparent, there being no obvious place where the measurements might be at fault.

CONCLUSIONS AND GENERAL DISCUSSION

This problem was primarily one of constructing apparatus and not one of obtaining data. The results obtained from the data taken, while definitely not of high accuracy, are, however, good enough to warrant the conclusion that the apparatus is fundamentally all right, and with further adjustment and refinement would undoubtedly render rather highly accurate results.

While the apparatus was still in the construction stage, the first ultrasonic waves produced were detected by the simple expedient of placing a thermometer in front of the acoustic crystal. Frictional heat at the liquid-glass and glassmercury boundaries due to the passage of the sound waves was sufficiently great to cause a five to ten degree rise in the thermometer reading in a very few minutes, while no effect was apparent outside the sound beam. This is one more evidence of the rather high concentrations of energy in an ultrasonic beam.

It was interesting to note that with this particular apparatus liquids of density greater than one apparently could not be used. Tuning the acoustic crystal tank circuit had no effect - the r.f. milliammeter in the acoustic crystal circuit showed no current, and the whole effect was as if the crystal circuit were open. This effect might be explained by the fact that the piezo-electric qualities of a crystal are reciprocal - not only does the application of a charge to the faces cause a mechanical deformation, but a mechanical deformation causes the appearance of charges on the faces. If

the charges are such as to cause an expansion, a pressure on the crystal will cause the appearance of similar charges. Now let us suppose that the phase of the r.f. current is such as to cause an expansion. At the same time the liquid is exerting a damping force on the crystal, which is probably some function of the density of the liquid. This would tend to produce a positive charge on the same face as the current would be producking a positive charge, and there would be in effect an opposing emf which would be a function of the damping force and thus a function of the density of the liquid. This opposing emf could be of such a magnitude as to balance out the applied emf, thus accounting for the observed results. This effect showed up in carbon tetrachloride and glycerine, and also in some liquids whose density was less than one - for instance, it was well-nigh impossible to set up a sound field in methyl alcohol, density 0.7921 at 23.8°C.

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