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ULTRASONIC PROPAGATION IN LIQUIDS

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ULTRASONIC PROPAGATION IN LIQUIDS:
I. Application of Pulse Technique
to Velocity and Absorption
Measurements at 15 Megacycles

by

J. R. Fellam and John Galt

Abstract

Equipment developed by the M.I.T. Radiation Laboratory has been applied to the measurement of sound velocity and absorption in liquids at 15 mc/sec. Pulses of one microsecond duration are generated by a transducer, which also picks up the resultant echoes from a plane reflector. Velocity measurements are made by determining the distance the transducer must be moved to delay the received echoes by a specified increment. Absorption measurements are made by determining the attenuation necessary to keep the receiver signal constant as the transducer is moved. The attenuation factor can be measured to an accuracy of about 5 percent and sound velocity to about 0.05 per cent. Measurements in homologous series of organic liquids are reported.

Title page

14 Numbered pages

I. INTRODUCTION

The electronic pulsed-circuit techniques and equipments developed during the war are adaptable to the measurement of velocity and absorption of sound in liquids¹. Preliminary work of this nature took place at the M.I.T. Radiation Laboratory², and the pulse principle has also been applied³ to the examination of mechanical flaws in metals. For velocity determination in liquids the pulse method is at least as accurate as optical diffraction and acoustic interferometer methods previously employed⁴, and considerably more accurate for absorption measurements. It is direct and convenient and therefore particularly adapted to investigations involving large numbers of liquids. The present paper reports measurements of sound velocity and absorption in selected organic liquids at 15 mc/sec and at several temperatures.

The scheme is essentially to use the liquid sample as a "storage medium" for short sound pulses and to measure the time delay and attenuation undergone by the sound in traversing a known path within the liquid. The acoustical pulses are generated from electrical pulses by means of a crystal transducer and are converted back to electrical form upon completing a transit through liquid; the effects of changing the acoustical path-length can be compensated for electrically. Hence the increased delay produced by an increase in path-length gives a measure of sound velocity; the attenuation which must be removed from the electrical circuit to balance acoustical losses in the additional distance provide a measure of absorption.

The variation in acoustical path-length is accomplished by mounting the crystal transducer on a movable support riding within a tank containing a sample of the liquid. The 15 mc/sec. sound pulses, after being generated from electrical pulses by the transducer, travel through the liquid to a plane reflector and back again to re-excite the crystal at a later time. The resulting delayed electrical signal passes through a calibrated attenuator to a receiver, the output of which is fed to an oscilloscope equipped with a special sweep

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- 1 This has been reported at the April 1946 meeting of the American Physical Society: Fellam, J. R. and Galt, J. K., abstract A12.
 - 2 Cefola, M., Droz, M. E., Frankel, S., Jones, E. M., Maslach, G., Teeter, C.E., Jr., Radiation Laboratory, M.I.T. Report 963, March, 1946.
 - 3 Firestone, F. A., Metal Progress 48, 505 (1945). Simon, E. N., Metal Progress, 48, 513 (1945).
 - 4 Bergmann, L., Der Ultrashall, (Edwards Brothers Reprint, Ann Arbor, Michigan, 1944).

indicating time delays; the start of this sweep is synchronized with the generation of the original pulse, and calibrated accurately with respect to time. The change of the signal position of the screen as the transducer mount is moved with respect to the reflector is a direct measure of the corresponding delay; simultaneously, attenuation is inserted or removed by means of the calibrated attenuator to keep the receiver input signal constant.

Velocity is obtained directly from the slope of distance traveled plotted vs. pulse delay, and absorption is obtained from the slope of compensating electrical attenuation vs. distance. Since the method depends only upon observing the effects of differences in acoustical path-length, knowledge of exact distance travelled by the sound is unnecessary.

Results obtained in this manner may be regarded as physically equivalent to those obtained by continuous wave methods, since it may be shown that differences in behavior between pulsed sound and continuous sound in liquids should produce negligible effects. For example, no significant dispersion in liquids has been detected at these frequencies⁴ (nor any pulse distortion during these measurements) so that the group velocities measured by pulsing should not differ measurably from the phase velocities ordinarily dealt with. Similarly, although attenuation of sound in liquids varies as the square of the frequency, the bandwidth associated with the pulses used is small enough to prevent appreciable effect on absorption measurements.*

II. EQUIPMENT

a). Electrical Equipment and Circuits

Fig. 1 is a block diagram of the equipment and Fig. 2 is a photograph of the assembled apparatus. The timing sequence is as follows: A crystal controlled circuit in the oscilloscope starts the scope sweep and simultaneously sends a trigger to the pulse generator. The pulse formed by the generator passes through a simple impedance matching network to the transducer; when the resulting sound pulse returns to the transducer after its round-trip within the liquid, the delayed electrical pulse is formed which passes back through the matching network. Since the transmitter is off when the echo signal arrives, very little leakage loss occurs and the main portion travels through the attenuator to the receiver.

* The exponential pressure attenuation coefficient (α), defined by

$$p = p_0 e^{-\alpha x}$$

will have a fractional error given by

$$\frac{\Delta \alpha}{\alpha} = \left(\frac{\Delta \nu}{\nu}\right)^2 \frac{2^2 \alpha}{\alpha^2 \nu^2} = \left(\frac{\Delta \nu}{2\nu}\right)^2$$

if measured over a frequency range of width (4). The last term results from placing (α) proportional to frequency squared, as is frequently the case. It may be shown that, for pulses of much longer duration than the period of a single oscillation, $\Delta \nu / 2\nu$ is just the reciprocal of the number of cycles per pulse. The pulses employed in the measurements reported here contained about 15 oscillations, corresponding to an error of roughly one part in 250.

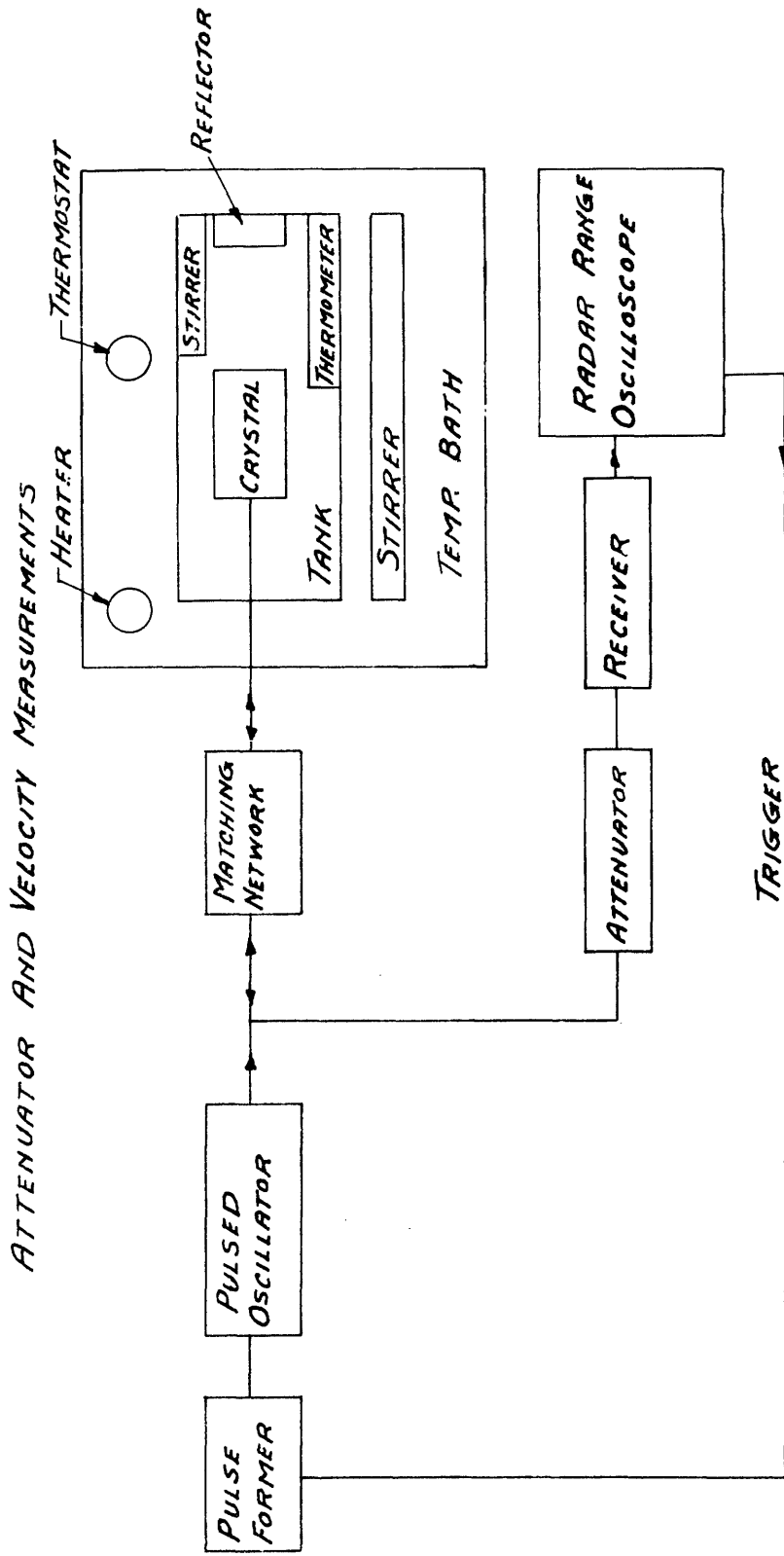
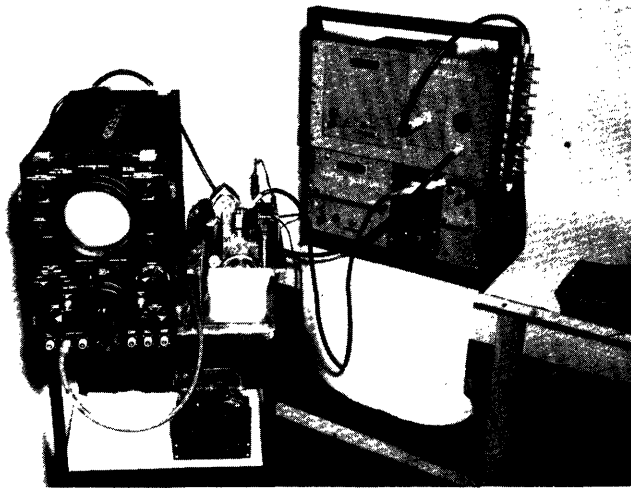


Fig.1 BLOCK DIAGRAM



Apparatus: left, A/R range scope; center, liquid delay tank; right, pulse forming network, oscillator, attenuator, and receiver

Fig. 2

All triggering and time measurements are accomplished by the crystal-controlled circuit in the oscilloscope (DuMont Type 256B A/R Range Scope), so that synchronization occurs automatically. In addition to setting off the pulse generator and starting the sweep, this circuit also provides marker pips spaced accurately every 12.192 microseconds thereafter (corresponding to 2000 yards radar range). Minute examination of the front edge of the echo signal is possible by means of an expanded sweep with variable delay and a speed of 1 inch per microsecond. Crystal control is necessary primarily for timing the marker pips accurately; one pip is developed for each oscillation, and since the crystal frequency is accurate to better than 1 part in 10,000 the same is true for the range marker positions.

A detailed discussion of the design of the pulse generator circuit has been given elsewhere.⁵ The trigger from the oscilloscope passes through a stage of amplification to excite the second stage, a blocking oscillator. The output of the blocking oscillator, after being clipped to form a video pulse, is applied through a switch tube to the 15 mc/sec. r.f. oscillator. The latter is driven into oscillation for the duration of the pulse, and the resultant r.f. pulsed signal feeds through two stages of amplification. The final signal level put out to the transducer may be controlled by means of a variable power supply to the amplifier.

Since the electrical impedance presented by the crystal is very different from the 70 ohm characteristic impedance of the transmission line leading from the pulse generator, an impedance matching network⁶ is provided to improve the response of the transducer. A small coil is used both as an autotransformer to match the line and as a means for tuning out the dead capacity of the crystal to give a purely resistive load (see Fig. 3). A coil of about 3 microhenries total inductance with a 6 to 1 turns ratio fulfills both requirements.

The receiver includes: (1) four stages of amplification at 15 mc/sec.; (2) a 15 mc/sec. local oscillator and mixer; (3) two stages of amplification at 30 mc/sec. (each stage consisting of three tubes with circuits tuned to slightly different frequencies for broad banding); (4) a detector; (5) two stages of video amplification; (6) a video amplifier in the oscilloscope. The 30 mc/sec. stages were used only because of equipment availability; a receiver operating at 15 mc/sec. throughout is now under construction.

The calibrated attenuator has steps of 1db. and up. Shielding is adequate to prevent leakage from introducing appreciable errors, even though attenuations as high as 110 db. are inserted.

5 Frankel, S., M.I.T. Radiation Laboratory Report 645-8,

6 The authors are grateful to Messrs. Jones and Frankel of the Radiation Laboratory for the design constants used in this network.

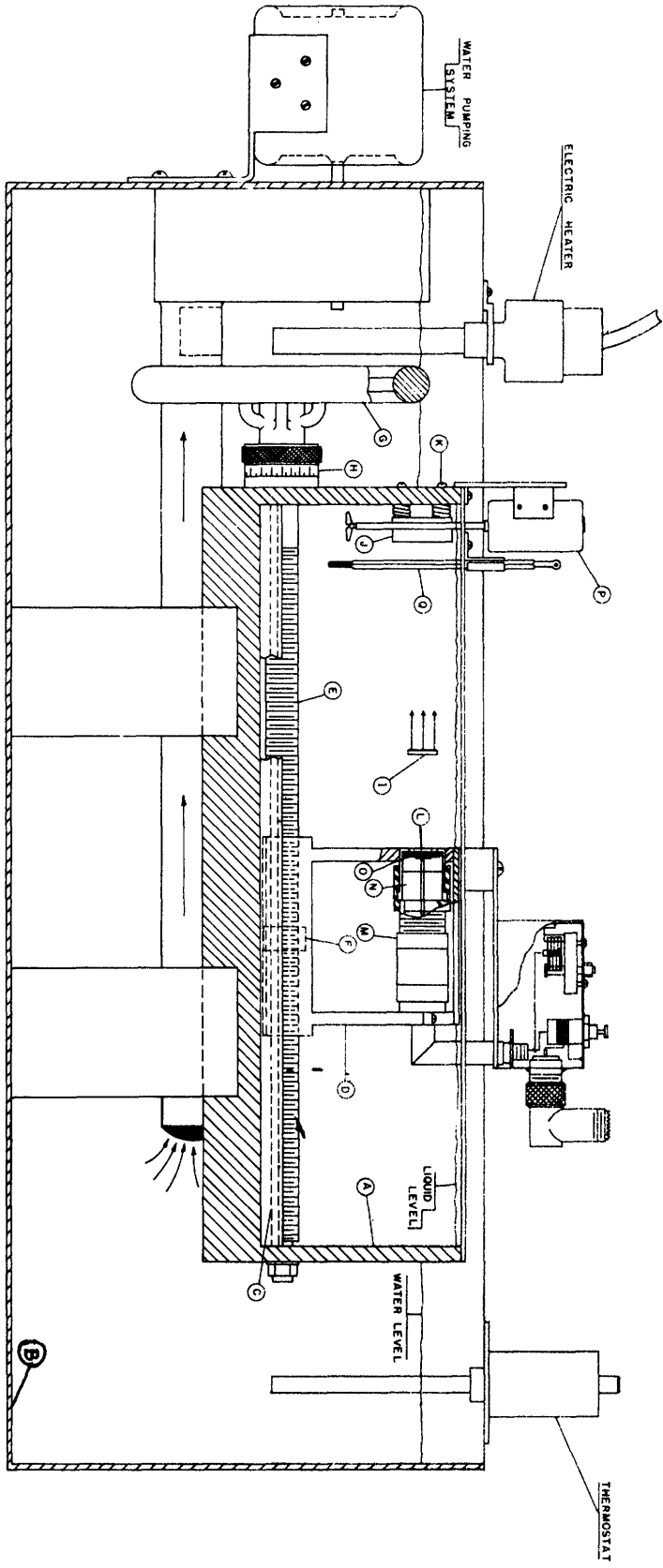


Fig. 3 LIQUID DELAY TANK

b). Sound Delay Tank for Liquids

A sample of the liquid being investigated is placed in a sturdy brass tank (A) (see Fig. 3) within a constant temperature bath (B). The bottom of the tank is sufficiently thick (1/2") to accommodate a milled track (C), with adjustable side plate for tightening, and upon which the transducer mount (D) travels. Motion is produced by turning the phosphor bronze micrometer screw (E) which threads through a stainless steel nut (F) set in the transducer mount. A hand-wheel (G) is provided and a dial (H) which indicates travel in thousandths of an inch. Calibration of the screw against size blocks makes it possible to read the transducer position significantly to 0.0001".

The sound pulse (I) is reflected at the end of the tank by a disk (J) of stainless steel, ground and lapped optically flat. This reflector is supported by three adjustment screws (K), and may thereby be set perpendicular to the sound beam by aligning for maximum returned signal strength.

The transducer element (L) is an X-cut 15 mc/sec. quartz crystal plate, 1/2" in diameter and gold plated on both sides. This crystal is held in place within a cartridge (M) by a lucite bead (N) so that a diameter of about 1 cm is exposed to the liquid. A metal foil (O) between the bead and the quartz establishes electrical contact with the back surface.

c). Temperature Control

Temperature equilibrium is maintained by balancing the effects of dry ice in the bath against the warming action of a thermostatically controlled electric heater. A water pumping system improves temperature uniformity outside the measuring tank, and a motor-driven stirrer (P) maintains constant temperature within the liquid sample itself. Temperature is read by thermometer (Q) and must be maintained to within less than 0.03°C for velocity measurements accurate to 0.01%.

III. MEASUREMENT TECHNIQUES

The velocity of sound within most organic liquids is of the order of 1.5×10^5 cm/sec. which corresponds at this frequency to about 100 wave-lengths per cm. Hence the crystal radiating surface is about 100 wave-lengths across so that the pulses travel out within a very sharp beam (0.7° to the 1st. minimum). It may be shown that this spread produces less than 1/2 db. of geometrical attenuation over the range employed; therefore readings of attenuation are suitable, without corrections for computing liquid absorption coefficients directly.

Velocity readings are taken at 2000 yard (radar) range intervals by placing the returned signal pip at definite positions with respect to successive scope range markers, and recording the setting of the micrometer screw. By properly adjusting the scope these markers appear as very short (dark) breaks in

the sweep, and if consistent positioning criteria are adopted, accurate range difference readings result. It turns out that careful adjustment of attenuation is necessary for consistent settings. Consequently simultaneous readings for velocity and attenuation are preferable.

This interdependence between measurements exists because the signal pulses never build up instantaneously, even though they are sharp; as a result the front edge of the pulse may actually be shifted with respect to the range marker by varying the attenuation level. Therefore always to the same arbitrary height on the screen (chosen within the linear range of scope amplification) the identical portion of the signal may be obscured by successive range markers. Once this condition is satisfied, the crystal position may be set more precisely if a predetermined attenuation is then arbitrarily removed; this allows setting on a lower, and therefore steeper, portion of the pulse front. Consistent results are obtained in this manner providing the same quantity is removed for each range reading, and a value of 10 db. was found convenient.

It is evident that if this adjustment of the returned signal to a predetermined level is attained by means of the attenuator leading to the receiver (see Fig. 1) without disturbing the gain of the remainder of the system, absorption data follows as a by-product of the velocity measurements.

For accurate absorption readings the sharpness of the beam requires that (1) the reflector disk be adjusted carefully, and (2) temperature gradients within the liquid sample be avoided. The proper operation of the stirrer (P) is absolutely essential to the temperature uniformity required for accurate absorption measurements. If the stirrer is stopped there is an immediate change in signal level and non-linear absorption curves result. Unless dispersed by circulation, the heat developed by attenuation within the beam produces these refraction effects.

IV. RESULTS

a). Linearity of Absorption Curves

Curves of attenuation vs. range are given in Fig. 4 for three sample liquids indicating the degree of linearity found (over a range exceeding 100 db. for CCl_4). No effects attributable to geometrical divergence of the beam are evident; the limit of accuracy appears to depend upon the smallest attenuation increment used (1 db.) as suggested by the small systematic variation about a straight line.

b.) Summary of Measurements

Part I of this paper is concerned only with the presentation of measurements in the form of raw data, interpretation being reserved for Part II. However, these results are given in terms of quantities or parameters possessing physical significance in view of Part II.

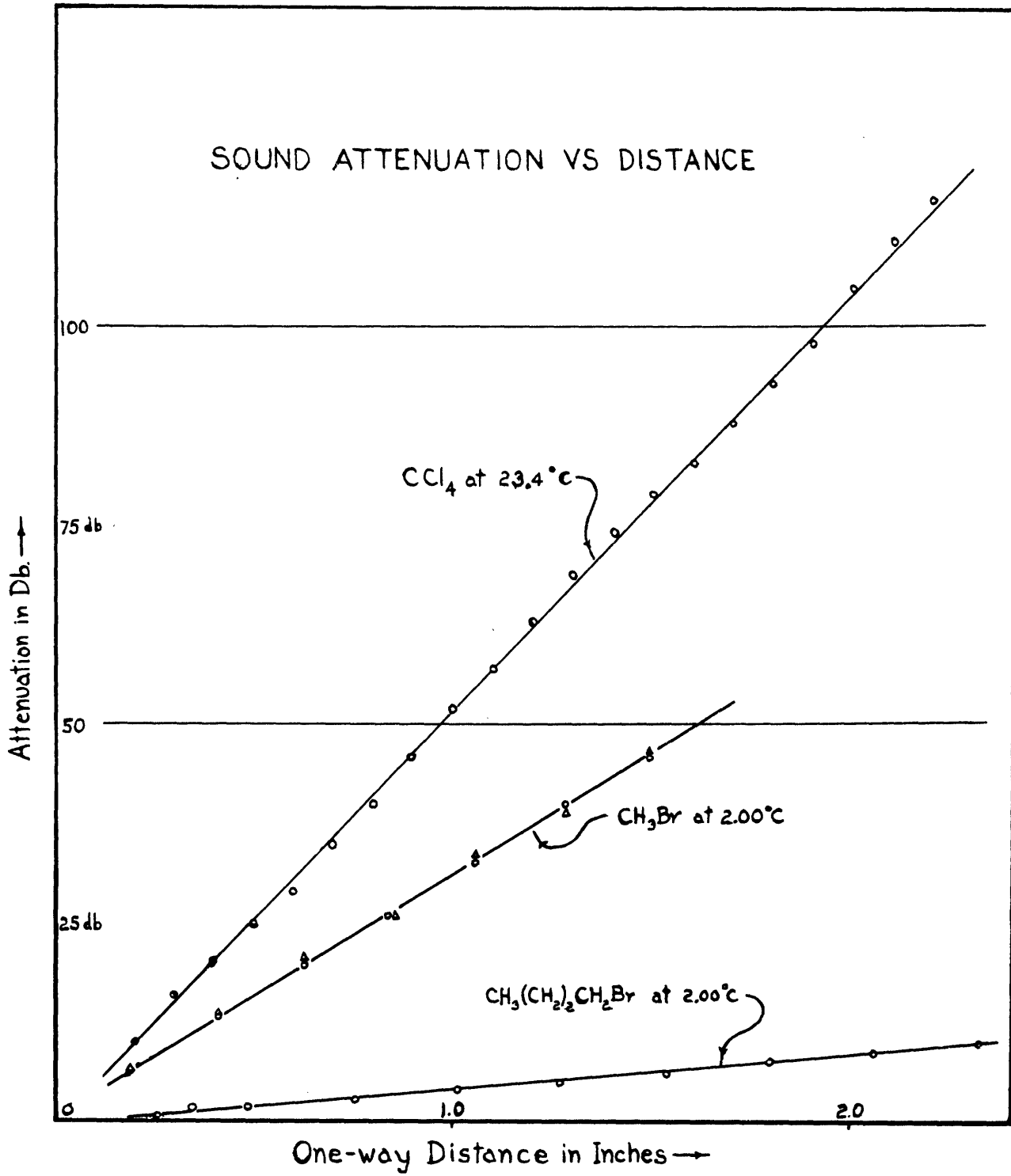


Fig. 4

Table I is a summary of measurements obtained for the various series of related organic compounds investigated. The velocity is given in Column (2) in units of cm/sec. and the absorption coefficients (α) in Column (3) in units of cm^{-1} . (α) is the exponential pressure attenuation coefficient defined by

$$p = p_0 e^{-\alpha x} \quad \text{Eq. 1}$$

Since for a number of liquids (α) varies nearly as the frequency squared, a convenient form for comparing measured results with those computed from classical (viscosity) theory is given in Column (4). The classical absorption values were calculated considering viscosity effects alone; heat conduction will increase these values by about 5%. Columns (6) and (7) give the experimental temperature coefficients for (α) and velocity, respectively.

These particular series of homologous compounds were chosen with a view to observing the gross effects of substitutions within molecules (such as the revelation for example that replacing an -H in Benzene by a -Cl cuts absorption by a factor of 5, and that an -NO₂ cuts it by 9). The most complete array examined was the Alkyl Halide group, of which 10 members were included (the 2° C was chosen just below the boiling point of Methyl Bromide).

c). Accuracy

Velocity results are given here to an accuracy of about 1 part in 1000. It is believed, however, that the pulse method is inherently capable of yielding as high accuracy for velocity measurements as the best diffraction techniques. On the other hand, whereas previous techniques were often incapable of exceeding 25% accuracy in absorption measurements, the absorption is given here to about 5% on the average. Since the limit of this accuracy is set by the smallest attenuation increment best results occur for highly absorbing liquids since they provide a greater attenuation range. Thus for liquids such CCl (α) is given to about 1%, but is accurate only to about 10% for liquids exhibiting low absorption.

d). Temperature Dependence of Absorption

The pulse method for measuring (α) is sufficiently accurate for investigating the effects of temperature. For example Fig. 5 shows the absorption coefficient for Methyl Iodide plotted against temperature (for values up to the boiling point). Temperature coefficients may be obtained from such curves and, as shown in Part II of this paper, have considerable significance with regard to the mechanism of sound propagation.

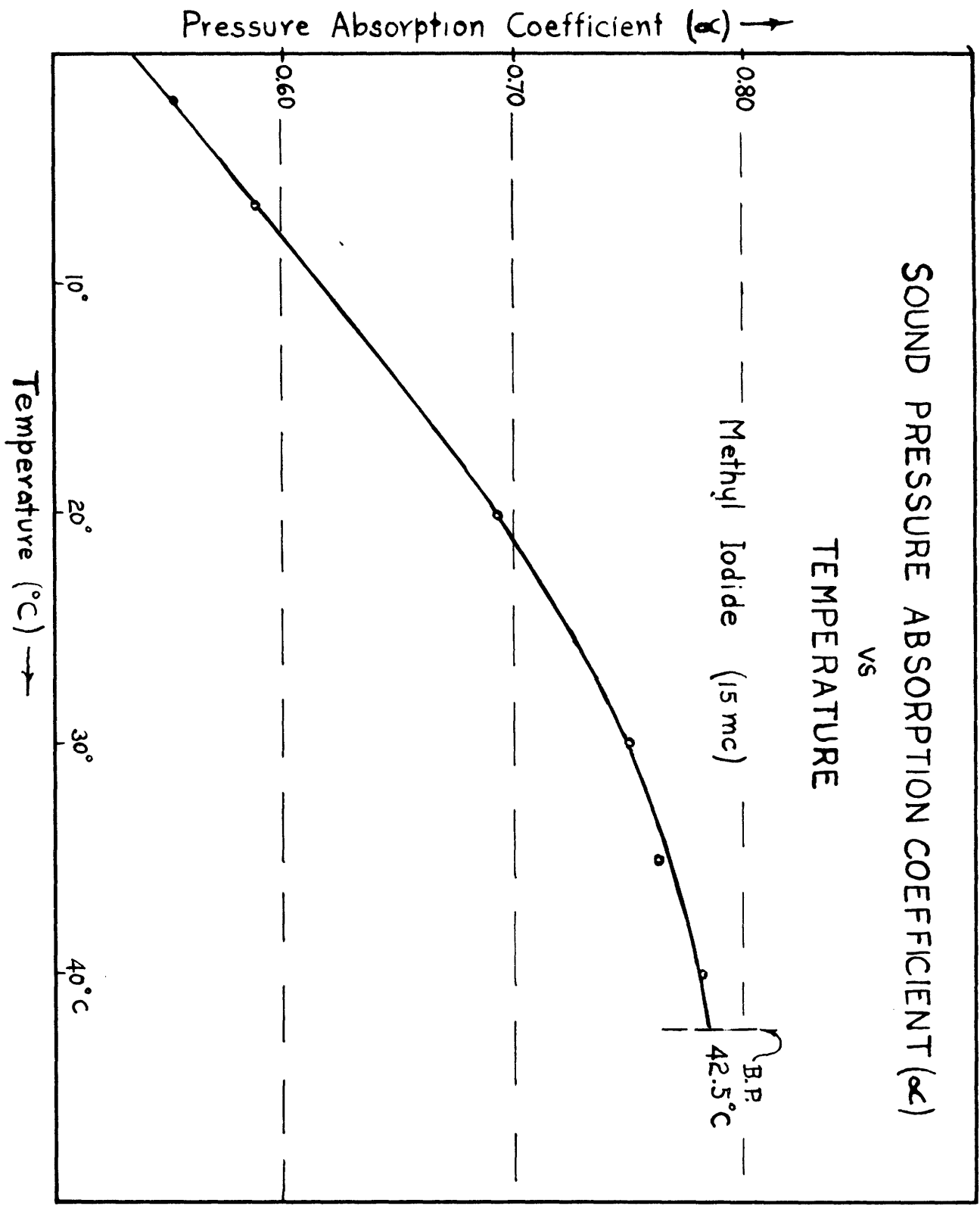


Fig. 5

Table I
Experimental Values of the Pressure Attenuation and Velocity in Liquids

Frequency $f = 15$ mc/sec.

$$P = P_0 \exp(-\alpha x)$$

Substance	(1) Temp. °C	(2) Velocity cm/sec.	(3) Attenuation (α) cm ⁻¹	(4) (a/f^2) $\times 10^7$ sec ² /cm	Experimental		(7) $\frac{d\alpha}{cdT}$ °C ⁻¹
					Classical	Exper.	
Methyl Bromide CH ₃ Br	2.0	0.905 $\times 10^5$	0.694 (+1%)	304	--	--	--
Ethyl Bromide CH ₃ CH ₂ Br	2.0	0.959	0.136 (+5%)	61	10	0.010	-0.0036
	10.0	0.932	0.14	62			
	20.0	--	0.14	62			
	30.0	--	0.18	80			
n-Propyl Bromide CH ₃ CH ₂ CH ₂ Br	2.0	1.0326	0.088 (+5%)	39	11	--	--
n-Butyl Bromide CH ₃ (CH ₂) ₂ CH ₂ Br	2.0	1.083	0.11 (+10%)	49	13	--	--
Methyl Iodide CH ₃ I	2.0	0.888	0.555 (+1%)	247	10	+0.0087	-0.0031
	6.5	--	0.588	262			
	20.0	--	0.711	316			
	30.0	0.815	0.751	334			
	35.0	--	0.755	340			
	40.0	--	0.776	345			
Ethyl Iodide CH ₃ CH ₂ I	2.0	0.9235	0.09 (+10%)	40	12	--	--
n-Propyl Iodide CH ₃ CH ₂ CH ₂ I	2.0	0.982	0.122 (+5%)	54	14	--	--
	2.0	1.022	0.109 (+5%)	48	17	--	--
n-Butyl Iodide CH ₃ CH ₂ CH ₂ CH ₂ I	2.0	1.158	0.095 (+10%)	42	8	--	--
	2.0	1.213	0.242 (+2%)	108	10	--	--

Table I (con't.)

Substance	(1) Temp. °C	(2) Velocity cm/sec.	(3) Attenuation(α) cm ⁻¹	(4) (α/r^2) $\times 10^{17}$ sec ² /cm	(5)		(7) $\frac{d\alpha}{cdT}$ °C
					Classical	Exper.	
Methyl Acetate CH ₃ COCH ₂ (Fract)	19.0	1.195	0.16 (+3%)	71	6	—	—
Ethyl Acetate (Fract) CH ₃ COC ₂ H ₅	24.3	1.145	0.163 (+3%)	72	8	—	—
Amyl Acetate (Fract) CH ₃ COC ₅ H ₁₁	29.2	1.173	0.167 (+7%)	74	14	—	—
Methyl Alcohol CH ₃ OH	2.0 19.2	1.186 1.123	0.10 (+15%) 0.084 (+10%)	45 37	15 14	—	-0.0032 -0.011
Ethyl Alcohol CH ₃ CH ₂ OH	2.0	1.246	0.052 (+10%)	23	29	—	—
n-Propyl Alcohol CH ₃ CH ₂ CH ₂ OH	2.0 27.5	1.288 1.1976	0.195 (+2%) 0.157 (+3%)	87 70	55 36	—	-0.0028 -0.0086
n-Butyl Alcohol CH ₃ (CH ₂) ₂ OH	2.0	1.324	0.240 (+5%)	54	65	—	—
n-Amyl Alcohol CH ₃ (CH ₂) ₃ CH ₂ OH	2.8 28.6	1.320 1.224	0.348 (+2%) 0.231	155 106	113 58	—	-0.014 -0.0029
Benzene C ₆ H ₆	7.5 21.8	1.382 1.3116	1.52 (+3%) 1.79 (+1%)	680 800	9 8	—	-0.0036 +0.011
Nitrobenzene NO ₂ C ₆ H ₅	7.5 23.8	1.518 1.462	0.165 (+5%) 0.178 (+4%)	73 79	16 14	—	-0.0023 +0.0048
Chlorobenzene C ₆ H ₅ Cl	7.5	1.333	0.276 (+1%)	123	10	—	—
n-Hexane CH ₃ (CH ₂) ₄ CH ₃	21.2	1.085	0.174 (+5%)	77	10	—	—
n-Heptane CH ₃ (CH ₂) ₅ CH ₃	22.4	1.150	0.18 (+10%)	80	10	—	—
Carbon Tetrachloride CCl ₄	23.5	0.924	1.20 (+1%)	533	20	—	—

V. REMARKS

a). Anticipated Measurements

A new apparatus for measuring velocity and attenuation by the pulse technique has just been completed. It is designed for operation at liquid helium temperatures. Preliminary measurements have already been made for liquid nitrogen, and measurements in the liquified form of the inert gases will shortly be under way.

b). Acknowledgements

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