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Sound Absorption and Velocity in Liquefied Argon, Oxygen, Nitrogen, and Hydrogen*

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The pulse method has been applied to measure the velocity and absorption of sound in liquefied argon, oxygen, nitrogen, hydrogen at 44.4 Mc/sec. The experimentally determined attenuation (α) is compared with the calculated attenuation due to viscosity (α_{vis}) and the results indicate that relaxation processes are not an important mechanism for sound absorption in any of these gases, except perhaps for hydrogen. Measurements were also made at 14.8 Mc/sec., but the attenuations were too low for satisfactory precision. Indications, however, are that the attenuation varies as the frequency squared.

I. INTRODUCTION

MEASUREMENTS on pulses of ultrasonic vibrations have been used during and since the war to determine the absorption and velocity of sound in liquids.^{1,2} Absorption is determined from the attenuation of the pulse as it traverses longer and longer paths in the liquid; velocity is determined by the increase in time required for the pulse to traverse the longer distances in the liquid. The oscilloscope presentation units developed for fire-control radar during the war render such measurements quite simple.

The present research is the application of this method to measurement of ultrasonic absorption and velocity in liquefied argon, oxygen, nitrogen, and hydrogen. The primary effort has been placed on obtaining attenuations, since, to the author's knowledge, these have not been measured previously. Velocity measurements have been made with a precision of approximately 1 percent. Measurements have been made at 14.8 and 44.4 Mc/sec., but the attenuations at 14.8 Mc/sec. are so low that quantitative measurements could not be made with satisfactory precision. For this reason no values for attenuation are presented at 14.8 Mc, but indications are that the attenuation varies as frequency squared.

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¹ M. Cefola, M. E. Droz, S. Frankel, E. M. Jones, G. Maslach, and C. E. Teeter, Jr., M. I. T. Radiation Laboratory Report 963, March 1946.

² J. R. Pellam and J. K. Galt, *J. Chem. Phys.* **14**, 608 (1946).

II. EQUIPMENT

The electronic equipment used is essentially the same as that used in previous work on liquids² in this laboratory, and no further description of it will be given here.

The tank containing the liquid sample traversed by the ultrasonic vibrations³ is designed to fit into a helium cryostat of the Collins type.⁴ An isometric view of the equipment in place in the cryostat is shown in Fig. 1. A quartz crystal (*A*) is used as both the transmitting and receiving transducer. An optically polished fused quartz reflector (*B*) is used to reflect the transmitted pulse back to the quartz crystal. The reflector surface may be adjusted to be parallel to the quartz by rods (*C*), which run up above the top of the cryostat. The reflector is connected to a micrometer screw (*D*) at the top of the equipment by means of a fused quartz rod (*E*). Fused quartz is used in order to minimize errors caused by the thermal expansion of this rod in reading the position of the reflector. A small stirrer (*F*) driven by a motor (*G*) is used to minimize temperature gradients in the liquid sample. The electrical signal is fed in by means of a coaxial cable (*H*) through the matching network. This equipment has proved quite satisfactory for the present research, but a later model developed by Pellam and Squire⁵ is more suitable for very low temperature work.

³ This equipment was designed in collaboration with J. R. Pellam.

⁴ S. C. Collins, *Rev. Sci. Inst.* **18**, 157 (1947).

⁵ J. R. Pellam and C. F. Squire, "Ultrasonic velocity and absorption in liquid helium," M. I. T. Research Laboratory of Electronics Technical Report No. 44, August 1, 1947; *Phys. Rev.* **72**, 1245 (1947).

III. TEMPERATURE CONTROL, MEASUREMENT

Refrigeration for the argon and some of the oxygen runs was obtained by placing the lower end of the tank (see Fig. 1) in a large Dewar flask and then pouring liquid nitrogen and liquid oxygen into the Dewar. The equilibrium temperature of such an arrangement is determined by the proportions of liquid nitrogen and oxygen used. The level of the liquid in this outer bath was always well above the sample on which measurements were made in order to minimize the temperature gradient over the ultrasonic path. For this same reason the outer bath was continuously stirred.

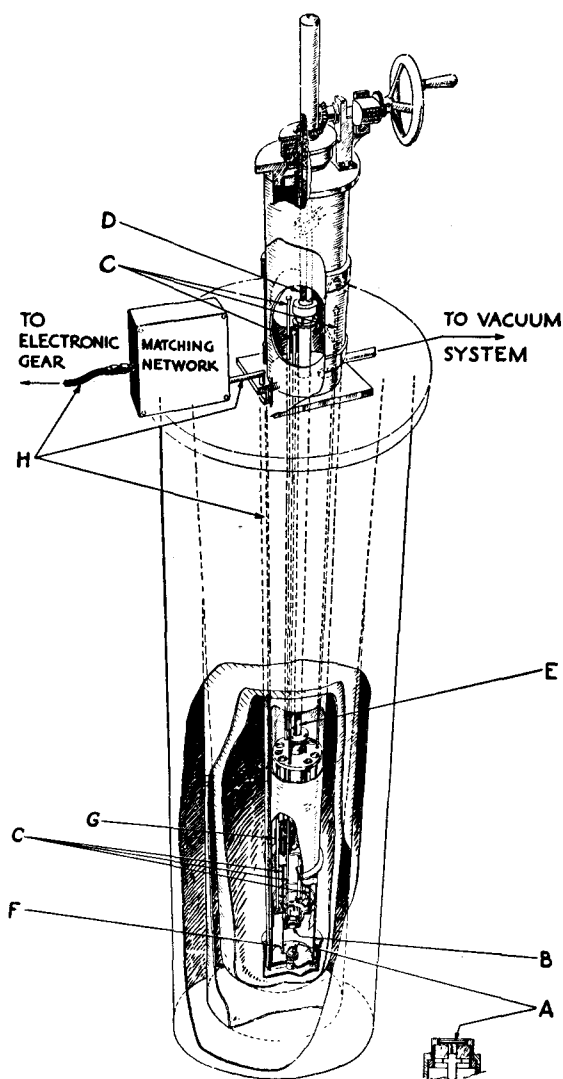


FIG. 1. Low temperature ultrasonic tank.

For the lower temperature measurements on oxygen, and for nitrogen and hydrogen the Collins cryostat was used to obtain refrigeration, as the oxygen-nitrogen mixture would not produce low enough temperatures. Holding the temperature constant is somewhat more of a problem with this equipment, but readings were taken quickly enough so that only on one oxygen run was the variation with time appreciably more than one degree.

Temperatures were measured in two ways. First, the system was closed off and the vapor pressure of the liquid was measured with a mercury manometer. Second, a constant-volume helium thermometer in the cryostat was used. The vapor pressure measurement was satisfactory except in the case of one run on oxygen. Here a small amount of nitrogen impurity raised the vapor pressure above that of oxygen alone. The helium thermometer was quite satisfactory, but it is not a direct measure of the temperature in the liquid sample, of course. No large differences between the two thermometers occurred, fortunately, except in the case of the oxygen run mentioned above. Here there is an uncertainty of about 5°K in the temperature reading.

IV. PURITY OF SAMPLES

The purity of the gases from which the liquid samples used for these measurements were obtained was stated by the supplier (Air Reduction Company) to be as follows: argon 99.8 percent with 0.2 percent nitrogen; oxygen 99.5 percent with 0.5 percent nitrogen; nitrogen 99.5 percent with 0.5 percent oxygen; hydrogen 99.5–99.7 percent with 0.3 to 0.5 percent oxygen. No water was present in any of the gases. As an additional precaution the hydrogen was put through a charcoal trap immersed in liquid nitrogen before it was used.

The impurity in the nitrogen may have caused some change in the velocity of sound, since the value measured here agrees with that of Hirschlaff⁶ rather than that of Liepmann⁷ (see Table I). It should be mentioned, however, that the freezing point was not appreciably different from that for pure nitrogen.

⁶ E. Hirschlaff, *Proc. Camb. Phil. Soc.* **34**, 296 (1938).

⁷ H. W. Liepmann, *Helv. Phys. Acta* **11**, 381 (1938); **12**, 421 (1939).

V. RESULTS

The results are shown in Table I. Attenuation α is defined by the relation $p = p_0 e^{-\alpha x}$ where x is the distance between the point where the peak pressure due to the sound wave is p_0 and the point where it is p .

The velocity measurements are accurate to within 1 percent. However, as mentioned previously, the value for nitrogen may have been influenced by the impurity of the sample. The value for hydrogen also differs somewhat from the published value,⁸ but it is felt that the present value is the more accurate in this case.

On the basis of the reproducibility of the data and the degree to which they fit the assumed exponential dependence on path length, an estimate of the maximum errors in the attenuation may be made. This leads to an estimate of 10 percent or less for argon, hydrogen, and oxygen, and 15 percent or less for nitrogen. Temperature fluctuations in the liquid, which in turn cause fluctuations in the signal, are the most important limitation on accuracy in reading signal amplitude. This effect is minimized by the stirrer, but it is not eliminated.

In theory, the total attenuation which we measure is due to viscosity, heat conductivity, and relaxation phenomena in the liquids.⁹ The attenuation to be expected from viscosity (α_{vis}) is presented in Table I. The densities used in these calculations are obtained from Bergmann.¹⁰ The viscosities were obtained from a compilation by Nason.¹¹ No data on heat conductivity could be found for these liquids, so it has been impossible to take this factor into account. The data would seem to indicate that relaxation processes are not an important mechanism for sound absorption in any of these liquids, pos-

TABLE I. Ultrasonic absorption and velocities.
(All values are measured at 44.4 Mc.)

Liquid	Temp. (°K)	Velocity in units of 10^4 cm/sec.	α_{exp} (cm ⁻¹)	α_{vis} (cm ⁻¹) calculated at 44.4 Mc
Argon	85.2±0.2	8.53	0.20	0.16
Oxygen	87.0±0.2	9.52	0.17	0.11
	70 ±1	10.94	0.17	0.11
	60 ±5	11.19	0.17	0.17 at 60°K 0.12 at 65°K
Nitrogen	73.9±0.2	9.62	0.21	0.13
Hydrogen	17 ±1	11.87	0.11	0.01

sibly with the exception of hydrogen. Here the α_{vis} value is not so good as in the other cases since the viscosity at the boiling point was the only value available, and the measurements were made at a lower temperature.

In view of the relatively small difference between the measured α and α_{vis} in nitrogen, any impurities present can hardly have caused a large variation in the attenuation.

No appreciable variation of attenuation with temperature was observed. This is especially noticeable in the case of oxygen. Measurements made within a degree or so of the boiling point of any of these liquids indicate an increased attenuation (attenuations of 0.27 cm⁻¹ have been measured in nitrogen), but this is attributed to the presence of bubbles and cavitation caused by the ultrasonic wave.

A magnetic field of the order of 1000 gauss was applied to the oxygen sample in our experiment to see whether this would produce a change in attenuation.¹² The experimental arrangement was such that a change of 1 percent in velocity or 20 percent in attenuation could have been observed, but no variations were noted.

VI. ACKNOWLEDGMENTS

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¹² A. Van Itterbeck and L. Thys, *Physica Haag* **5**, 298 (1938).

⁸ A. Pitt and W. J. Jackson, *Can. J. Research* **12**, 686 (1935).

⁹ L. Bergmann, *Der Ultraschall* (Edwards Brothers, Inc., Ann Arbor, Michigan, 1944), p. 191 ff.

¹⁰ Reference 9, p. 171.

¹¹ W. C. Nason, "Some physical properties of air and its components," compiled for Aero-Medical Laboratory, Wright Field under Contract No. W-33-038. His references for viscosities are: Rudenko and Schubnikow, *Phys. Zeits. Sowjetunion* **6**, 470 (1934); Rudenko, *J. Exper. a. Theoret. Phys. U.S.S.R.* **9**, 1078 (1939).