ABSORPTION OF ULTRASONIC WAVES BY VARIOUS GASES

By T. P. Abello

Abstract

Absorption coefficient for ultrasonic waves in CO_2, N_2, H_2 and He.—A beam of ultrasonic waves with a frequency of 612 kilocycles from an oscillating quartz crystal was passed through a mixture of gas and air, contained in a brass absorption tube with ends covered by thin celluloid films. The design of the tube greatly reduced the effect of resonance and allowed the use of two different lengths of absorbing gas, leaving the geometrical arrangement of openings and absorbing screens the same. The emergent beam was allowed to fall upon another quartz crystal which had a natural frequency very nearly equal to the frequency of the waves and which was connected to the grid of a triode. The intensity of the received signal was measured by a previously calibrated vacuum tube voltmeter. A logarithmic decrease in the transmitted energy with increase in the percentage of the gas in the mixture was found with CO_2, N_2O, H_2, and He. The increase in the absorption coefficient (cm^{-1}) at 612 kilocycles when air was replaced by a mixture containing 1 percent by volume of the gas was found for CO_2 to be 0.029; for N_2O, 0.034, for H_2, 0.014, for He, 0.0025. For argon mixtures no absorption was observed.

Reflection of ultrasonic wave by a thin celluloid film. The fractional part of the beam which was transmitted by the films at the ends of the absorption tube, increased with increase in percentage of CO_2 and N_2O but decreased with increase in percentage of H_2 and He, agreeing qualitatively with Rayleigh's theory.

Introduction

IN THE course of his measurement of the velocity of ultrasonic waves in carbon dioxide, Pierce[1] noticed that at a frequency of 1,034 kilocycles the gas was practically opaque to the waves. Even at 98 kilocycles he noticed a much greater absorption in carbon dioxide than in air. The writer has observed the decrease of the transmitted intensity with increasing percentage of the gas for 612 kilocycle waves from a quartz crystal. In the first experiments the ultrasonic beam was passed through a mixture of carbon dioxide or hydrogen and air contained in a brass tube with ends covered by the celluloid films. The intensity of the emergent beam was measured by the pressure it exerted against a torsion vane and it was found that there was a nearly logarithmic decrease in the intensity with an increase in the percentage of the gas.[2] The experiments have been continued with a new form of absorption tube which greatly diminished the resonance effect and which allowed the determination of the absorption curves for two different lengths of tube with the same films at the ends.

Apparatus

The arrangement of the apparatus is shown in Fig. 1. C_1, the source of the waves, was a piezo-electric quartz crystal, connected to an oscillating

[1] G. W. Pierce, Proc. Am. Acad. Arts & Sci. **60**, 298 (1925).
[2] T. P. Abello, Proc. Nat. Acad. Sci. **13**, 699 (1927).

circuit as shown. The crystal radiated waves with a frequency of 612 kilocycles as determined by the electrical frequency of the oscillator. The oscillator was completely shielded except for the window O, of about 2 cm diameter, in the metal wall W. Emerging from O, the waves passed through the absorption tube A, the ends of which were covered by thin celluloid films, then through one of the holes, also covered with film, in the metal disk D, and finally struck the face of the receiving crystal C_2. The pressure of the waves induced an alternating potential of the same frequency as the incident waves on the opposite faces of the crystal. This signal was impressed on the grid of the first tube. It was then heterodyned, amplified and, by a second transformer, applied to the grids of the last two tubes which were connected in parallel. The negative grid bias on these tubes

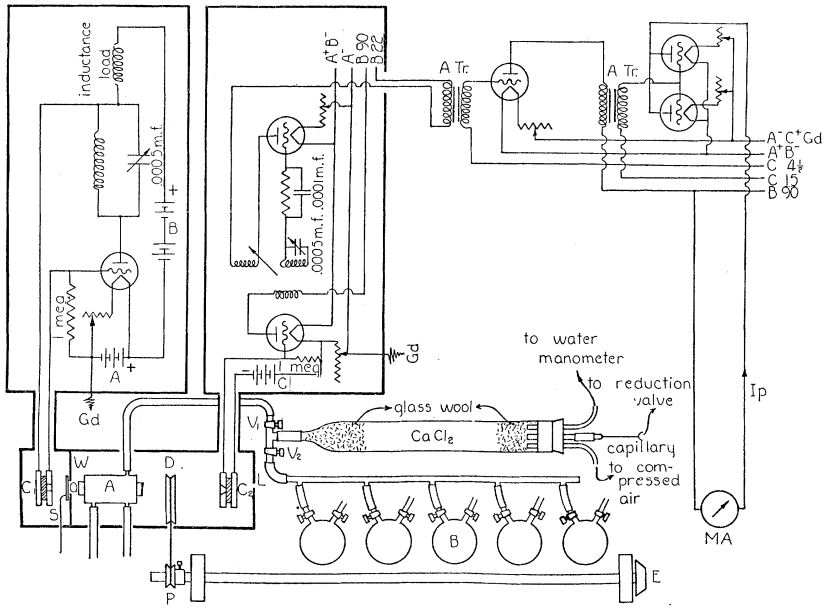

Fig. 1. Diagram of apparatus.

was so adjusted that they were operating near the foot of the I_p-E_g characteristic curve, so that when O was closed by means of the bakelite shutter S thus stopping completely the waves, the milliammeter MA read nearly zero. They thus acted as a peak voltage voltmeter. All the tubes (UV 201A) were mounted on cushions and the metal shields were supported on corks to absorb vibrations. By means of two brass stopcocks V_1 and V_2 the mixture of the gas and air, coming from the $CaCl_2$ tube, could be passed either through A or through any of the glass balloons B. A ten-liter bottle was inserted between the compressed air pipe and the drying tube to keep the flow of air constant. The composition of the mixture was altered by changing the pressure in the reduction valve which was placed before a capillary.

Crystal and mountings. The two crystals which were cut from the same natural crystal, were of the same shape and very nearly the same thickness. They were ground to have nearly the same natural frequency. It was found that they vibrated more strongly when the opposite faces were polished with fine rouge. In the experiment the waves were propagated in the direction of the electric axis. The crystals were held between brass electrodes and by increasing the pressure on the crystal its frequency could be varied within a small range. The electrodes had circular holes, about 1.5 cm in diameter, opposite the center of the crystal, through which the waves passed. For the receiving crystal the edges of the hole were bevelled at about 45° to prevent reflected waves striking the crystal.

The absorption tube. The absorption in various gases was observed by introducing them into a tube through which the sound passed. The ends were closed by thin celuloid films, so that the two crystals always oscillated in air,

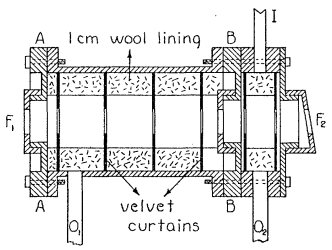

Fig. 2. Absorption tube.

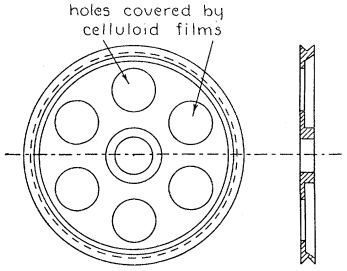

Fig. 3. Calibration disk.

no matter what gases were being studied. Fig. 2 shows a section of the absorption tube used. It was a brass tube having the inside lined with a 1 cm layer of wool. Along the entire length, placed at nearly regular intervals, were velvet curtains which limited the beam to a diameter of 1 cm. The holes in the caps F_1 and F_2 which were held on by wax, were covered with thin celluloid films formed on a clean mercury surface. To test whether the film was air-tight, the cap was inverted in a vessel and enough water was poured in until the confined air was under about 0.5 cm difference of pressure. The film bulged out a little and if the bulging remained for several hours the film was considered sufficiently tight. The film at F_2 was placed not exactly normal to the direction of the beam but inclined at a small angle. In that position reflection from it was towards the velvet curtains and wool linings where it would be dissipated by multiple reflections, instead of towards the other film. This form of tube greatly reduced, although it did not completely eliminate, resonance. This is important for since the wave-length inside the tube changed with the change in the composition of the mixture while the length of the tube was constant, the tube would be in resonance for certain mixtures but not for others. The tube was made so that it could be used to give two different lenths of absorbing gas leaving the geometrical arrangement of openings and absorbing screens the same.

This was done by interchanging the sections AA and BB. The new position of the film F_1 made a tube length of only 3.5 cms for the gas mixture which entered at I and left at O_2. In the arrangement shown the tube length F_1F_2 was 10 cms and the gas mixture entered at I and left at O_1. To make the joints air-tight, rubber washers were placed between sections.

The calibration disk. The brass disk D used to determine the intensity transmitted was about 9 cm in diameter. It had six circular holes 1.5 cm in diameter, with their centers equally spaced on a circle 5 cm in diameter. The holes were covered with celluloid films of different thicknesses, formed on a clean surface of water. These films were very much thinner than those at the ends of A. The two thinnest were white while the others showed interference colors. They were, however, very porous and so could not be used to cover the ends of A. As shown in Fig. 3, the edge of D was grooved so that by means of a pulley and rod it could be rotated from E (Fig. 1). In this way one could place in succession the various films in the path of the beam and at the same time read the milliammeter MA. The transmitting powers of these films were previously determined so that the reading of MA could be expressed in terms of the intensity of the beam striking the face of the receiving crystal.

Determination of Transmitting Powers of Films

It was thought at first that the transmission through these films could be obtained from Rayleigh's formula, and their thicknesses were therefore determined by an interferometer. It turned out however that the transmission through them as measured by a torsion vane was very much less than the value computed from Rayleigh's formula. It was therefore decided to use the transmission, as measured by the torsion vane, in this work. This was done in the following way. Referring to Fig. 1, the absorption tube A and the receiving crystal C_2 were removed from the box, the inside of which was lined with a 0.5 cm layer of wool, to absorb waves striking it. At the same position occupied by the face of the receiving crystal, a very thin circular mica vane of the same size as the hole in the electrode was suspended, with its plane vertical, from a graduated torsion head by a quartz fiber 0.002 cm thick and 15 cm long. Fig. 4 shows how the vane was suspended. The suspension and vane were completely shielded from air currents except for the two small holes through which the string, used to rotate D, passed. The amount of rotation which was required to restore the vane to its original position was taken as a measure of the intensity of the beam. All the holes were covered with films, the transmission through the thinnest that could be made being about 30 percent of the transmission through an uncovered hole.

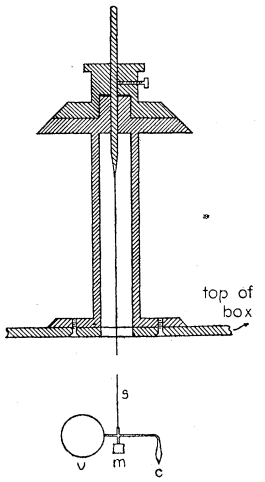

Fig. 4. Torsion vane.

Different series of measurements gave very consistent values for the amount transmitted by the various films. Taking the transmission through the thinnest as 100, the values for the others are 79.1; 52.6; 42.1; 29.6 and 7.8.

Measurement of the Absorption

In making an absorption measurement, the curve connecting the milliammeter readings with the intensity of the waves striking the face of the receiving crystal was first taken by placing the various films in the path of the beam. The disc D was left with the thinnest film in the path of the beam. Fig. 5 shows such a calibration curve which we will refer to as an I_p-Intensity curve; the intensity transmitted by the thinnest film with air in the absorption chamber being arbitrarily taken as 100.

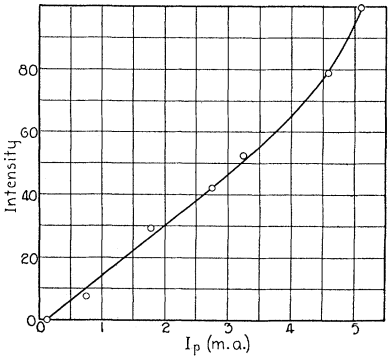

Fig. 5. I_p-Intensity calibration curve.

A little gas was then allowed to mix with the air. The milliammeter reading dropped slowly and after a while, generally about 1.5 minutes, when the mixture flowing through A had become constant, as indicated by the constant reading of MA, the readings of MA, the reduction valve, and the water manometer were recorded. Then the rate of flow of the gas was increased by increasing the pressure in the reduction valve and the above process was repeated. Superimposed on the gradual decrease in the ammeter reading with increasing proportion of added gas, there was a fluctuation of small amplitude caused by the recurrence of resonance as the mixture reached certain proportions. This effect was most prominent with hydrogen and helium and was hardly noticeable in carbon dioxide and other gases with high absorption. In a run five different mixtures were used. After the readings for the last mixture were taken, the flow of the gas was stopped and MA was read again for pure air to be sure that the circuits had not changed during the interval (generally about 12 minutes). If the run was good, sections AA and BB of the tube A were interchanged, the I_p-Intensity curve was determined again for this new setting, and the above procedure was repeated for the shorter tube using the same percentage mixtures as those which had been used with the longer tube. Finally samples, corresponding to the different mixtures used, were taken. To do this the valve V_1 was closed and V_2 was opened and the mixture was allowed to pass through one of the glass balloons, B. The reduction valve was adjusted so that it read the pressure used before and by means of V_2 or the inlet valve of the glass balloon, the rate of flow of the mixture was regulated so that the water manometer gave the corresponding reading. The bulbs containing the samples of the mixtures were then weighed on an analytical balance and the volume percentage of the mixture was determined from the I_p-Intensity curve.

It is evident that when the percentage of the mixture inside A is changed, the fractional part of the beam which is reflected from the films also changes. It was in order to eliminate this correction experimentally that the absorption curves for two different lengths were determined. If x is the percentage (by volume) of the gas in the mixture, there is a certain function $f(t,x)$ of x and the thickness t, which gives the fractional part of the energy transmitted by the films at the ends of the absorption tube. In the tube the absorption coefficient of the mixture $\mu(x)$ depends on x. The intensity at the receiving crystal, I_{1x} with a tube of length l_1 will depend on x according to a formula of the type $I_{1x}=Af(t,x)e^{-\mu(x)l_1}$. The experiments agree with the relation $\mu(x)=\mu(0)+kx$, giving for the dependence on x,

$$I_{1x}=I_0 f(t,x)e^{-kl_1x} \tag{1}$$

where I_0 is a constant. The coefficient $k=[\mu(x)-\mu(0)]/x$ is the difference in the absorption coefficient of pure air and of a mixture containing 1 percent by volume of the gas studied.

If the length of the tube is increased to l_2, the transmitted intensity with the same mixture and the same films is $I_{2x}=I_0 f(t,x)e^{-kl_2x}$. By division we get

$$I_{2x}/I_{1x}=e^{-klx} \text{ or } \log (I_{2x}/I_{1x})=-klx$$

where $l=(l_2-l_1)$ is the difference in the length of the two tubes. Therefore by dividing the ordinates of the absorption curve for the longer tube by the corresponding ordinates of the curve for the shorter tube (intensity is the ordinate) the effect of reflection at the gas surfaces could be eliminated, and what we obtain is the true absorption curve due to the difference in length. From this curve the value of k is readily determined. Moreover we see that once the value of k is known we can also learn something about the dependence of $f(x,t)$ on x. For from (1) we get

$$I_0 f(t,x)=I_{1x}e^{kl_1x}$$

giving the dependence of the transmission function $f(t,x)$ on the percentage of gas in the mixture.

Results and Discussion

Carbon-dioxide and nitrous-oxide. These two gases showed exactly similar behavior (Figs. 6 and 7). Both absorbed the waves very highly and for them the function $f(t,x)$ increased with x, which means that the fractional part of the incident waves which was transmitted by the films increased as the percentage of the gas in the mixture was increased. The coefficient k in (1) may be deduced from the curves. When the length is in centimeters, and x is the percent by volume, k for CO_2 is 0.029 and for N_2O is 0.034.

Hydrogen and helium. The curves (Fig. 8 and 9) obtained for hydrogen and helium are similar although the helium curves show very much less absorption than those for hydrogen and there seemed to be a tendency for

the helium curves for the two different lengths of tube to come together at high percentage. But I would not emphasize this fact as the experimental error at low intensity was proportionately great. There is, however, a very

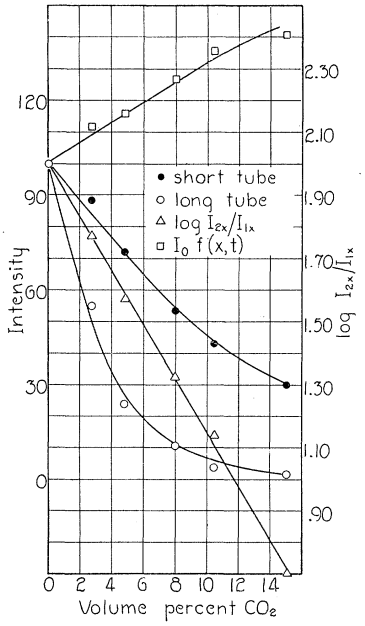

Fig. 6. Variation of intensity with volume-percent of CO_2.

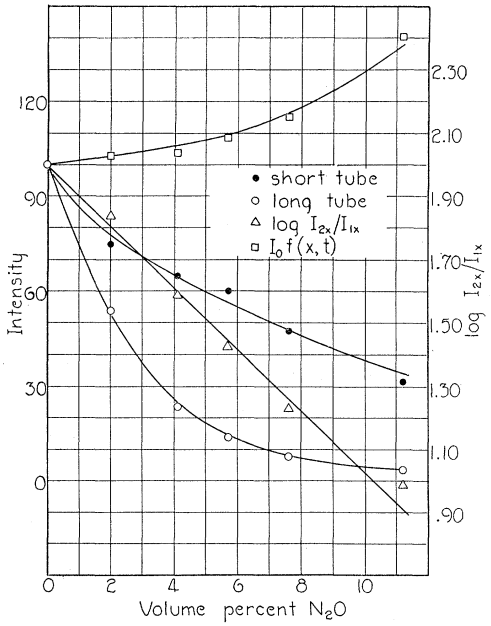

Fig. 7. Variation of intensity with volume-percent of N_2O.

striking contrast between the behavior of these two lighter gases and the behavior of carbon dioxide and nitrous oxide, namely, that for hydrogen and helium $f(t,x)$ is a decreasing function of x, while for the other two, it is

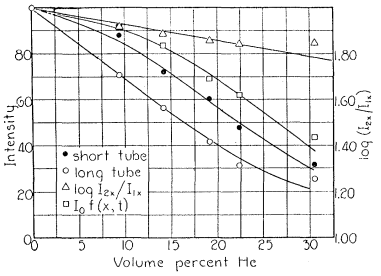

Fig. 8. Variation of intensity with volume-percent of He.

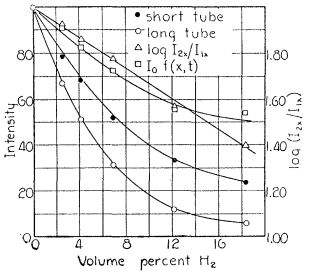

Fig. 9. Variation of intensity with volume-percent of H_2.

an increasing function. We shall come to this fact again later. In the case of helium this new method of determing the absorption curves for two different lengths of tube showed that much of the decrease in intensity

[3] T. P. Abello, Phys. Rev. **31**, 157 (1928).

previously reported[3] was due to the increase in reflection at the gas surfaces. The value of the coefficient k for H_2 deduced from Fig. 9 is 0.014 and for He deduced from Fig. 8 is 0.0025.

Argon. In experiments with this gas a slow current of argon was allowed to flow into the tube A thus slowly increasing the percentage of the mixture. Apparent absorption was noticed at certain concentrations, but on closing the inlet and outlet of A the absorption disappeared almost instantly, indicating that the apparent absorption was due to scattering caused by the inhomogeneity of the mixture. With pure argon the milliameter reading was the same as with pure air.

Ethylene. No quantitative measurement of the absorption in ethylene was made because the percentage of the mixture could not be determined by the difference in weight. However, this gas was also tried and a rough idea of its behavior was obtained by plotting the intensity against the pressure of the reduction valve. With the lean mixtures used in the experiment, the percentage of the gas would be nearly proportional to this pressure. From the relative slopes of the curves obtained for short and long tubes, it seemed as if the absorption in this gas is of the same order of magnitude as the absorption in CO_2 and N_2O.

Transmission by Films from a Rare to Dense Medium

Let us now return to the discussion of the observed behavior of the function $f(x,t)$. Extending the method used by Rayleigh to determine the energy of the transmitted and reflected waves at a thin infinite surface with the same medium on both sides, to the case of different media, we obtain the following expression for the reflected energy:

$$\text{Reflected energy} = \frac{\cot^2 a_2 l\left(\frac{\rho_3}{\rho_1}-\frac{a_3}{a_1}\right)^2+\left(\frac{v_1\rho_3}{v_2\rho_2}-\frac{v_2\rho_2}{v_3\rho_3}\right)^2}{\cot^2 a_2 l\left(\frac{\rho_3}{\rho_1}-\frac{a_3}{a_1}\right)^2+\left(\frac{v_1\rho_3}{v_2\rho_2}+\frac{v_2\rho_2}{v_3\rho_1}\right)^2} \tag{3}$$

where $a=2\pi/\lambda$, λ is the wave-length, ρ, the density, l, the thickness of the second medium, and v, the velocity of the waves. The subscripts 1, 2, and 3 refer to the first, second, and third medium respectively, In this experiment, l was the thickness of the film and since $l \ll \lambda_2$, $\cot^2 a_2 l = (\lambda_2/2\pi l)^2$ very approximately. Furthermore we can write

$$\frac{v_1\rho_3}{v_2\rho_2}-\frac{v_2\rho_2}{v_3\rho_1}=-\frac{v_2\rho_2}{v_3\rho_1}$$

very approximately since $v_1v_3/v_2^2 \ll \rho_2^2/\rho_1\rho_3$. Introducing these approximations, the reflected energy becomes

$$\text{Reflected energy} = \frac{\left(\frac{\beta-1}{2}\right)^2+\left(\frac{\pi l\rho_2}{\lambda_1\rho_1}\right)^2}{\left(\frac{\beta+1}{2}\right)^2+\left(\frac{\pi l\rho_2}{\lambda_1\rho_1}\right)^2} \tag{4}$$

where $\beta = v_3\rho_3/v_1\rho_1$. When the third medium is the same as the first medium the above formula reduces to Rayleigh's formula. (Theory of Sound, II, p. 88).

$$\text{Reflected energy} = \frac{(\pi l\rho_2/\lambda_1\rho_1)^2}{1+(\pi l\rho_2/\lambda_1\rho_1)^2} \tag{5}$$

Interchanging subscripts 1 and 3 in Eq. (3) we also find expression (4) for the reflected energy, showing that the fraction reflected is the same for waves incident in the first medium as for waves incident in the third medium and that the effect of our two films is the same as double tramsmission from air through the film into the mixture.

Now a close examination will show that Eq. (4) is greater than Eq. (5) for $[(2\pi l\rho_2/\lambda_1\rho_1)^2+1] < \beta < 1$ but is less than Eq. (5) for $[(2\pi l\rho_2/\lambda_1\rho_1)^2+1] > \beta > 1$. In the experiment β cannot be greater than $[(2\pi l\rho_2/\lambda_1\rho_1)^2+1]$. In other words the reflected energy will always be greater for any third medium satisfying the condition $\beta < 1$ than for the same medium on both sides. The reverse will be the case for the transmitted energy. Using the velocity of audible waves for lack of experimental data since very little has been done on the velocity of high frequency waves, we see that with air as the first medium, β is less than 1, when hydrogen, helium or their mixtures with air form the third medium. The opposite is the case with carbon-dioxide and nitrous-oxide. Hence the experimental curves for $f(x,t)$ agree at least qualitatively with Rayleigh's theory on the above supposition.

An increase of velocity in CO_2, as observed by Pierce for 206 kilocycles, would still further increase the theoretical transmission in CO_2. Hitchcock,[4] however, reported recently that the velocity of the waves from a vibrating crystal is a function of the energy which the crystal radiates, and he obtained a value in air nearly twice the velocity of ordinary sound waves. If this is the case, undoubtedly the velocity in other gases will also be very different from the velocity of ordinary waves. Experiments along this line are very desirable.

The writer wishes to take this opportunity to express his indebtedness to Professor A. J. Dempster who had suggested this investigation, for his valuable advice and criticisms throughout the progress of the work. He also wishes to thank Dr. Barton Hoag for helping him in cutting the crystals and Mr. Fred Pearson of the optical shop for the final grinding and polishing of them.

RYERSON PHYSICAL LABORATORY,
UNIVERSITY OF CHICAGO.
March, 1928.

[4] R. C. Hitchcock, Proc. Inst. R. E. **15**, 906 (1927).