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Speed of Sound in Several Liquids

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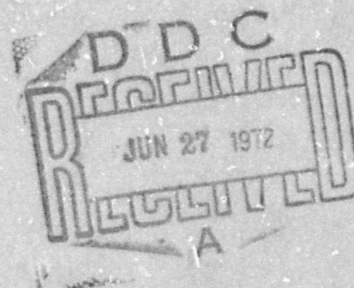
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Compatibility of materials

II

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Abstract

The speed of sound in several liquids has been measured as a function of temperature between 0 and 40°C. Data are given for DB castor oil (Baker Castor Oil Co.), FC-45 (a fluorinated hydrocarbon by 3M, Inc.), DC 200/.65 (a silicone fluid by Dow Corning Co.), Robane, Robalene, and Robuoy (organic hydrocarbon fluids by Robeco Chemicals Co.), octyl acetate (Eastman, Inc.), and Coolanol-35 (a Freon-type fluid by Monsanto). The compatibility of these liquids with the more commonly used transducer elastomers has been measured.

Problem Status

This is an interim report on one phase of the problem; work is continuing on this and other phases.

Problem Authorization

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SPEED OF SOUND IN SEVERAL LIQUIDS

Introduction

Liquids often are used as acoustic transfer mediums in underwater sound devices; therefore, the speed of sound in them is essential information for the equipment designer. Accordingly, the Underwater Sound Reference Division (USRD) has undertaken the measurement of sound speed as a function of temperature in a number of liquids likely to be used for this purpose. The temperature range of interest is that which might be encountered in the open ocean: 0 to 40°C.

The specific liquids investigated were DB castor oil (Baker Castor Oil Co.), FC-43 (a fluorinated hydrocarbon by 3M, Inc.), DC 200/.65 (a silicone fluid by Dow Corning Co.), Robane, Robalene, and Robuoy (organic hydrocarbon fluids by Robeco Chemicals Co.), octyl acetate (Eastman, Inc.), and Coolanol-35 (a Freon-type fluid by Monsanto). The first three fluids were studied because of an immediate need by the USRD for use in a reciprocity coupler. The last five were included because of possible applications to sonar equipment being developed for the Navy by Bell Laboratories. Data for several other liquids have been reported by Del Grosso [1].

Experimental Technique

Sound speed was determined by a time-of-flight technique in which the time required for a wavetrain to travel from a projector to a receiver along a straight-line path was measured. Projector and receiver are located at opposite ends of a high-pressure reciprocity coupler that has been described by Sims and Henriquez [2]. The two transducers consist of identical stacks of two square PZT-4 piezoelectric ceramic plates (1.9 × 1.9 × 0.635 cm) wired in parallel with a resonance frequency of 160 kHz. The projector is driven in a pulsed mode at the off-resonance frequency 400 kHz to take advantage of the shorter "ring" time of the crystal and the greater resolution that accompanies higher frequencies. Although still higher frequencies are advantageous for sound speed measurements, problems such as absorption that increases as the square of the frequency preclude their use with some liquids (castor oil, for example).

A block diagram of the experimental apparatus is shown in Fig. 1. A 400-kHz wavetrain eight cycles long is transmitted from the projector ceramic through the liquid in the coupler to the receiver ceramic. A transmit gate receives a start signal for the timer-counter from a pre-selected point on the transmitted wavetrain, and the receive gate ends the count upon receipt of the same preselected point. The time interval

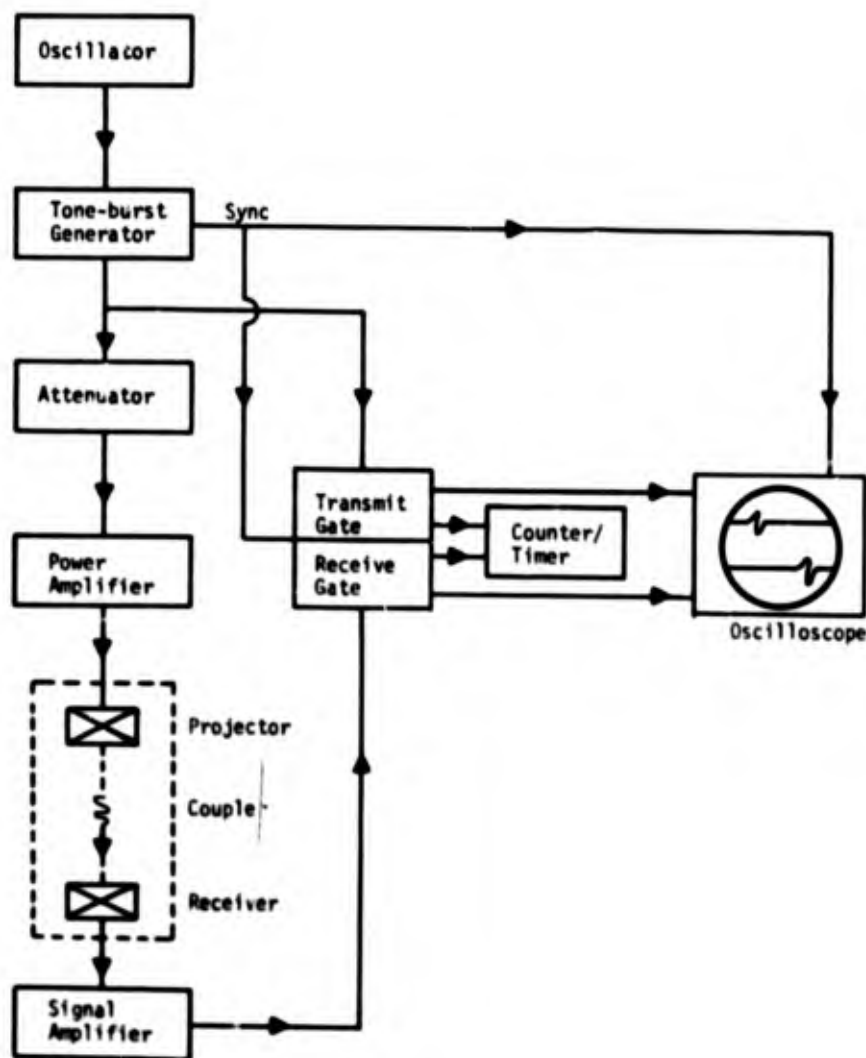


Fig. 1. Schematic of setup for measuring time of flight of sound pulse.

corresponds to the time of flight of the wave pulse between the two ceramic elements.

A thermistor that has been calibrated against a precision mercury bulb thermometer is positioned within the coupler to measure the temperature of the liquid at the edge of the actual flight path.

Confidence in the measured time of flight depends inherently on the degree of certainty with which the temperature and counter accuracies are known. Temperature can be determined to $\pm 0.5^{\circ}\text{C}$ and the counted time of flight to $\pm 0.03 \mu\text{sec}$. The uncertainty in temperature can be expressed as a percentage of sound speed once the temperature rate of change of the sound speed in the liquid is known. The rate of change of sound speed with temperature generally is different for different liquids, as will be shown in the next section; however, for the liquids under consideration

here, the error due to the uncertainties in temperature and counter always was less than $\pm 0.3\%$.

No attempt was made to measure the spacing between the projector and receiver to a limit compatible with the measurement accuracy of the temperature variations of time of flight. Instead, the coupler was filled with distilled water and the projector-to-receiver distance was calculated from the measured time of flight and known sound speeds in distilled water [3]. Because it was necessary to remove the projector and receiver for thorough clearing of the coupler between fluid changes, an additional uncertainty was introduced in that the projector-receiver spacing was not exactly the same for different fluids. Repeated measurements of the spacing after fluid changes indicated that the spacing was repeatable to ± 0.037 cm, which is equivalent to $\pm 0.6\%$ inaccuracy.

Thus, the relative sound speed in the liquids could be determined to $\pm 0.3\%$ and the absolute speed of sound to $\pm 0.9\%$.

Times of flight were measured in the temperature range 0 to 40°C at intervals of 5°C . Sound speed calculated directly from the measured time of flight differs from the true sound speed, however, because of dispersion due to viscosity. The results must be corrected by means of a formula given by Vigoureux [4]:

$$\frac{c}{c_0} = 1 - \frac{1}{r} \left(\frac{\mu}{2\rho\omega} \right)^2, \quad (1)$$

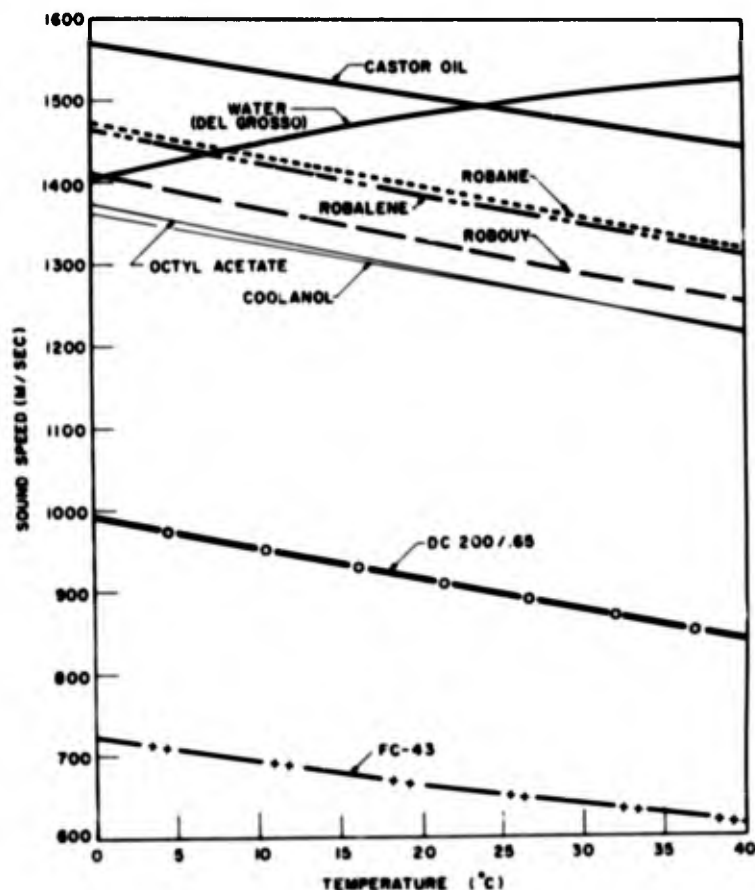
where c is the speed of sound in a closed tube of radius r , c_0 is the speed of sound in a free field, μ is the MKS unit of dynamic viscosity, ρ is the density of the liquid, and $\omega = 2\pi \times \text{frequency}$.

Dynamic viscosity and density were not known as a function of temperature for any of the liquids other than castor oil. Fortunately, however, the correction is small because of the size of the coupler ($r = 2$ cm) and the frequency (400 kHz). The effect of the small variation of density with temperature is completely insignificant in comparison with measurement uncertainties discussed above. Dynamic viscosity can vary over wide ranges, inversely with temperature and directly with pressure; but, again, the correction is small except for very large values of viscosity. Of the fluids studied, castor oil was the most viscous by about two orders of magnitude, with a viscosity of 7 P at 25°C . The dispersion correction factor, obtained from values of the viscosity given in the *International Critical Tables* [5], was applied to the time-of-flight data for castor oil. The correction factor was comparable to the relative sound speed uncertainty of $\pm 0.3\%$ only below 5°C . Hence, for the other less viscous liquids, any dispersion effects were negligible.

Experimental Results

The speed of sound in castor oil has been measured as a function of pressure as well as temperature and has been reported elsewhere [6]. The

Fig. 2. Sound speed in several liquids as a function of temperature.



pressure range was 0 to 110 MPa. The data were fitted with an empirical equation:

$$c(T,p) = 1570(a_0 + a_1T + a_2T^2 + a_3Tp + a_4p + a_5p^2) \pm 0.9\%, \quad (2)$$

where 1570 m/sec is the sound speed in castor oil at 0°C and atmospheric pressure, $a_0 = 1.000$, $a_1 = -2.15 \times 10^{-3}/^\circ\text{C}$, $a_2 = 4.0 \times 10^{-6}/(^\circ\text{C})^2$, $a_3 = 2.5 \times 10^{-6}/(^\circ\text{C} \cdot \text{MPa})$, $a_4 = 2.22 \times 10^{-3}/\text{MPa}$, $a_5 = -3.0 \times 10^{-6}/\text{MPa}^2$, T is temperature in degrees Celsius, and p is pressure in megapascals.

A comparison of the sound speeds in the various liquids as a function of temperature can be seen in Table I and Fig. 2. The sound speed in distilled water given by Del Grosso [3] has been included for comparison. Other characteristics of the liquids are presented in Table II.

Discussion

The sound speed in castor oil as a function of temperature has been measured previously by Del Grosso and Smura [1], and a comparison of data is presented in Fig. 3. The sound speed of 1580 m/sec at 0°C and 1 atm reported by Del Grosso and Smura is in agreement within experimental error achieved in the present measurements; however, the temperature

Table I. Comparison of speed of sound ($\text{m}\cdot\text{sec}^{-1}$) in several liquids as a function of temperature ($0-40^{\circ}\text{C}$).

Temp. ($^{\circ}\text{C}$)	DB					Octyl			FC-43	
	Distilled water	castor oil	Robane	Robalene	Robuoy	acetate	Coolanol-35	DC 200/.65		
0	1402	1570	1469	1464	1410	1373	1362	993	726	
5	1426	1553	1454	1444	1390	1352	1345	975	710	
10	1447	1537	1435	1423	1370	1331	1325	957	694	
15	1466	1521	1415	1404	1349	1311	1307	936	679	
20	1482	1505	1395	1385	1329	1291	1288	918	664	
25	1497	1490	1376	1366	1309	1272	1271	899	651	
30	1509	1474	1358	1349	1291	1254	1253	881	638	
35	1520	1460	1340	1331	1273	1236	1237	862	625	
40	1529	1445	1322	1314	1254	1218	1221	844	612	

Table II. Physical characteristics of several liquids.

	DB castor oil	Robane	Robalene	Robuoy	Octyl acetate	Coolanol-35	DC 200/.65	FC-43
Chemical formula	$C_{18}H_{34}O_3$	$C_{30}H_{62}$	$C_{26}H_{54}$	$C_{19}H_{40}$	$C_{10}H_{20}O_2$			
Pour point ($^{\circ}C$)	-23	-38	-35	-60	-39	< -85	-68	-50
Boiling point ($^{\circ}C$)	265	350	312	290	200		100	210
Sound speed* (m·sec ⁻¹)	1493	1380	1370	1313	1277	1276	902	627
Specific gravity*	0.957	0.809	0.806	0.780	0.868	0.890	0.751	1.87
Adiabatic compressibility* (m ² ·N ⁻¹ ×10 ⁻¹¹)	46.9	64.9	66.1	74.4	70.6	69.0	164	136
Viscosity* (P)	7.2	0.28	0.22	0.05	0.015	0.10	0.005	0.05
Resistivity* (Ω ·cm)	6×10^{12}	3×10^{14}	2×10^{14}	7×10^{13}	1.5×10^{10}	4×10^{10}	1×10^{13}	1×10^{14}
Temp. coefficient of sound speed* (m·sec ⁻¹ · $^{\circ}C^{-1}$)	-3.1	-3.7	-3.7	-3.8	-3.7	-3.4	-3.7	-2.8

*at 24 $^{\circ}C$.

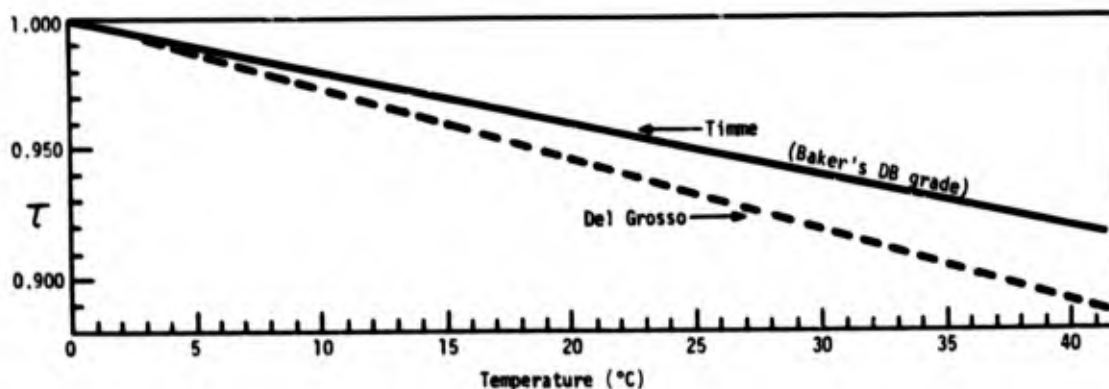


Fig. 3. Relative sound speed in castor oil as a function of temperature, normalized at 0°C and atmospheric pressure.

variation of the sound speed is not in agreement. A possible explanation for the discrepancy is that Del Grosso and Smura measured one of the Baker castor oils other than the DB grade. They identify the oil used in their measurements only as "Baker."

Sound speed measurements were made on samples of DB castor oil that had been carefully degassed and dehydrated by heating and stirring under vacuum and also upon samples that were untreated. The measured sound speed was the same in each case and independent of this treatment. Therefore, the sound speed measurements were made on the other liquids without special treatment.

The DC 200/.65 silicone fluid has been studied by Weissler [7] at a specific temperature and by Wilson [8] as a function of temperature. The data reported here are in good agreement with their results.

Robane, Robalene, and Robuoy are completely miscible with each other and with castor oil; thus, a mixture of these liquids could result in a number of fluids with sound speeds intermediate to those shown in Fig. 2.

Fluid Compatibility with Elastomers

When used as an acoustic transfer medium in sonar systems, a liquid necessarily is in intimate contact with other system components such as elastomers. It is as imperative that the liquid be compatible with the elastomers as it is to have the proper density and sound speed. Because of the question of compatibility between the liquids studied and elastomers, the results of a simple experiment are reported here.

The elastomers most commonly used in USRD underwater sound transducers are Type W neoprene G6470, NASL-H862A butyl, B252 butyl (all by Smithers Laboratories), and 35007 natural rubber (by B. F. Goodrich). The compound ingredients have been reported for the first three in a report giving the speed of sound in the elastomers [9]. Samples of these elastomers were immersed in the various liquids and physical properties such as size, weight, and ultimate tensile strength were monitored as a

Table III. The incompatibility factor (unit $\times 10^{-7} \text{ sec}^{-1}$) as a measure of the effect of various liquids on elastomers.

Elastomer	DB				Octyl acetate			
	castor oil	Robane	Robalene	Robuoy	Coolanol-35	DC 200/.65	FC-43	
Type W	0	0	0	0	31	0	4	0
H862A	0	19	12	19	34	19	12	0
B252	0	29	9	20	45	5	11	0
35007	0	34	28	17	109	26	23	0

function of immersion time. The measurements were performed at 25°C and were continued for 250 hours.

As used here, the term "incompatible" describes any liquid-elastomer combination that results in a physical change that would decrease the lifetime or effectiveness of a transducer. Such changes would include gain of weight by the elastomer or decrease in its tensile strength.

For certain pairs of liquids and elastomers, the elastomers underwent a marked amount of swelling, weight gain, and decrease in ultimate tensile strength. The changes in weight and tensile strength appeared to follow an exponential function of immersion time until saturation was approached. More detailed studies would be necessary to determine whether the changes are due to a chemical reaction or simply to a diffusion of the liquid throughout the elastomer. Because these liquids and elastomers are relatively inert, it is probable that diffusion of the liquid into the elastomer according to Fick's law results in a disturbance of the polymeric chain spacing and intermolecular forces.

For the purpose of comparing the effects of different liquids on the elastomers, the ultimate tensile strength (U.T.S.) was plotted against the natural logarithm of immersion time. The result is a straight line with a negative slope, indicating that the ultimate tensile strength obeys the relation

$$\text{U.T.S.} \propto \exp(-at) \quad (3)$$

where t is the immersion time and a can be defined as an incompatibility factor. Thus, the greater the value of a , the greater the effect of the liquid on the ultimate tensile strength of the elastomer and, by definition, the more incompatible the two.

In Table III, the incompatibility factor has been used to compare the effects of the liquids on the four elastomers. The results show that DB castor oil and FC-43 have the least effect on the elastomer, and octyl acetate is the most incompatible. Although it is readily agreed that the definition of the time rate of change of the ultimate tensile strength as an incompatibility factor is somewhat arbitrary, it serves a purpose by indicating that certain liquids are more desirable than others for use in transducers.

Acknowledgments

The samples of Robane, Robuoy, Robalene, octyl acetate, and Coolanol-35 were supplied by the Transducer Development Group of Bell Laboratories.

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