NRL Report 6567

Some Experiments with a Liquid Filled Acoustic Resonator Showing the Existence of Stable Bubbles in Water

A. J. Hiller

Techniques Branch
Sound Division

July 18, 1967

NAVAL RESEARCH LABORATORY
Washington, D.C.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ABSTRACT

Experiments were performed with a water-filled acoustic resonator to measure sound velocity in water as a function of temperature and undissolved gas. The measurements on gas-free distilled water agree well with theory, but measurements on sea water do not. This discrepancy can be accounted for by the existence of a stable microbubble population in the sea water, and by changes to this population as a result of the addition of thermal energy. Results showing that vacuum degassing can increase the sound velocity in the sea water sample demonstrated that a stable bubble population was present. Additional studies are needed for a clearer understanding of the nature of the stable bubbles and the role of particles in stable bubble formation.

PROBLEM STATUS

This is an interim report on the problem; work is continuing.

AUTHORIZATION

NRL Problem S01-26
SHIPSYSCOM Project SR 104-03-01-8136

Manuscript submitted March 16, 1967.

SOME EXPERIMENTS WITH A LIQUID FILLED ACOUSTIC RESONATOR SHOWING THE EXISTENCE OF STABLE BUBBLES IN WATER

INTRODUCTION

Acoustic scattering studies made in the Key West area (1) in January and March 1965 revealed the existence of particulate scattering layers at about 600 feet. Measured values of volume scattering strength were about 80 db higher than the values calculated using Rayleigh scattering theory. This large difference between measurement and theory can be accounted for by postulating that a multitude of submicron sized particles serve as bubble nuclei. To account for the persistence of the scattering layer it is also postulated that these bubbles are stabilized in some manner to give an average neutral buoyancy to the bubble system. The idea of a stable bubble probably originated with Harvey et al. (2), who postulated the trapping of air by hydrophobic particles. Later research by Fox and Hertzfeld (3) suggested stabilization by an organic skin on the inner surface of the bubble. Recent research by Turner (4,5) has given a more satisfactory hypothesis based on a continuous surface of fine particles. This is termed the "compressed wall hypothesis" and seems to be in agreement with experiments and observations by Liebermann (6) and by Turner.

Direct measurement of these sub-micron or micron sized bubbles is beyond the resolution of microscopic measuring methods. Sound velocity and attenuation are indirect methods sensitive to the presence of microbubbles and providing accurate measurements (7). The acoustic technique used in this investigation is based on sound modes propagated in a resonant chamber in which the resonant frequency is directly related to the sound velocity of the liquid in the chamber, and absorption is related to the decay constants.

APPARATUS

High Q spherical resonators (8) have been successfully used to measure relative absorption in the frequency range of 5 to 50 kc. The fundamental resonances of a liquid-filled thin-walled glass sphere are

$$\omega_n \approx \frac{n \pi C}{R} , \tag{1}$$

where n is the vibration mode, R is the radius, and C is the sound velocity of the liquid in the sphere. In the present investigation a simple Erlenmeyer Flask was used for the resonant chamber. The resonator and associated apparatus are shown in Fig. 1. It was determined experimentally that the first and second resonant vibration modes are determined by the relations

$$f_{01} = \frac{C}{2\ell} \tag{2}$$

$$f_{02} = \frac{C}{\ell} , \tag{3}$$

where ℓ is the peripheral length up to the water level.

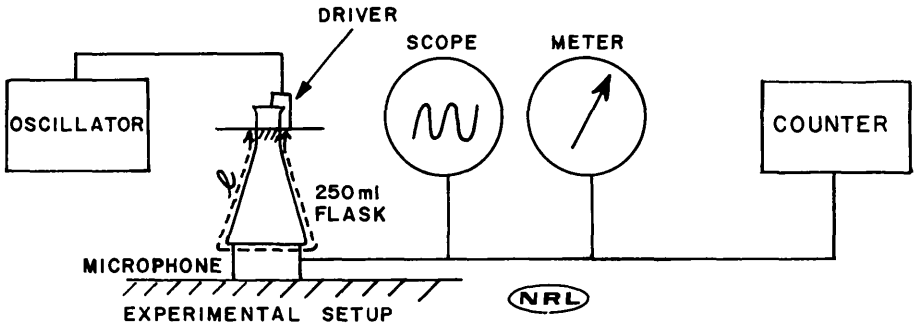

Fig. 1 - Experimental setup, where ℓ is the peripheral length up to the water level

Therefore, for a given path length ℓ and a given liquid sample in the resonator the sound velocity C is directly related to the resonant frequency. The second vibration mode was used in order to increase measurement precision and to permit mode excitation by the driver unit located at the lip of the flask. The bottom of the chamber is at a nodal point which conveniently allows the flask to be rested on the microphone. The resonant peaks are conveniently observed on the oscilloscope or the meter, and the resonant frequency is indicated on the digital counter.

The flask used to obtain the data in the following section was a 250-ml Erlenmeyer flask. The length ℓ was 1 foot, which conveniently makes the resonant frequency close to the actual sound velocity in ft/sec. For a given temperature, ℓ can be adjusted to give the "handbook" value of C_t by carefully adjusting the liquid level until the counter reads the value of C_t. However, this refinement is not necessary when comparing sound velocities, since the dimension ℓ cancels out. With care in observing the resonance peak, the resonant frequency can be measured to an accuracy of ±1 cps.

THEORETICAL CONSIDERATIONS

If the bubbles are much smaller than the wavelength of the sound, it can be shown (9) that the sound velocity is related to the fraction u of air in bubble form by the relation

$$C_0{}^2 = C^2 \left(1 + \frac{3u}{1.36^2 \times 10^{-4}}\right) \tag{4}$$

where C_0 is the velocity with no undissolved air present, and C is the velocity in the sample. Expressing u in parts per million it can be shown that

$$f = \frac{f_0}{1 + 0.0081u}, \tag{5}$$

where f_0 is the resonant frequency with no undissolved gas present, and f is the resonant frequency of the sample. The influence of undissolved air (bubbles) is shown in Table 1, which lists the frequency ratio f/f_0 and the air content in parts per million. Since the measurements are made with the same ℓ for a given sample, the velocity ratio C/C_0 is then identical with f/f_0.

EXPERIMENTAL RESULTS

The first experiments were conducted with fresh single distilled water produced by an all glass distillation unit. To determine the precision of measuring velocity ratios,

water with no microbubbles or a realizable minimum is desired. One set of measurements was made on the distilled water taken directly from the storage vessel (Table 2).

Table 1
Influence of Air Bubbles on the Frequency Ratio or Velocity Ratio

Air Content	Frequency Ratio or Velocity Ratio
u (ppm)	f/f_0 or C/C_0 *
0	1.000
1	0.992
2	0.984
3	0.976
4	0.969
5	0.961
6	0.954
7	0.946
8	0.939
9	0.932
10	0.925

*Using equation 5 approximation

Table 2
Measurements of the Resonant Frequency of Distilled Water

Temp (C°)	Resonant Frequency f (cps)
23.5°C	4783
28.5°C	4836

The measured frequency (or velocity) ratio is therefore

$$\frac{f_{28.5}}{f_{23.5}} = \frac{4836}{4783} = 1.01108$$

Velocity of sound-tables (10) for distilled water give the ratio

$$\frac{C_{28.5}}{C_{23.5}} = \frac{1506\ m/sec}{1493\ m/sec} = 1.00870$$

Thus the velocity table data gives a ratio 0.9976 of that obtained by the resonant frequency measurements.

The next experiment was conducted with a sample of Ocean City, Md., sea water collected near shore on 14 April 1966. The laboratory tests were made on 2 May 1966, giving over a two-week stabilization period. The data obtained is given in Table 3.

Table 3
Measurements of the Resonant Frequency of Sea Water

Temp (°C)	Resonant Frequency f (cps)
23.5°C	4780
31.0°C	4781

Velocity of sound tables for sea water (11) give the ratio

$$\frac{C_{31.0}}{C_{23.5}} = \frac{5070\ ft/sec}{5003\ ft/sec} = 1.01108$$

and the f ratios from the above measurements yield

$$\frac{f_{31.0}}{f_{23.5}} = \frac{4781}{4780} = 1.0002$$

The results of other tests are shown in Fig. 2. Also plotted are the published values (11) of sound velocity for distilled water and sea water of 32 ‰ salinity which is close to the value of Ocean City sea water. Since the resonant frequency is proportional to sound velocity, a direct comparison can be made between the slopes of the published curves and the slopes of the curves by measurement. It is noted that when the samples are degassed by vacuum (29 inches Hg) plus impact (sharp taps on flask) the slopes of the sea water and distilled water (curves D and E) are very close to the reference slopes (curves B and C). It is also noted that the slope of the degassed distilled water decreased when the temperature was raised, indicating that some dissolved gas may have been present to form bubbles. Aged distilled and aged Ocean City sea water (curves A and F) show the presence of bubbles by the decreased slopes of the curves.

DISCUSSION

It is important in resonant tests to establish that the measured effect is due to factors in the volume of the liquid in the test chamber and not due to a wall effect. This was established by using the f_{02} mode and probing the volume with a free gas probe, which was a small piece of sponge attached to the end of a swab stick. The air entrained by the sponge as it probed the flask showed that resonance was changed in the central volume region of the flask.

Clean glassware is essential, since nonwetting regions on the wall can collect gas. Dichromate cleaning followed by an alcohol rinse and additional distilled water rinses is a good procedure. Improper cleaning of glassware is detected by vacuum-impact degassing which results in vaporous cavitation occurring at a fixed place on the flask surface.

Degassing of sea water by vacuum alone is ineffective. Cavitation by impact to the flask wall causes profuse vaporous cavitation at nuclei throughout the liquid volume. Distilled water also contains numerous particles carried over in the distillation process and by normal atmospheric dust. The test for these particles is also impact cavitation.

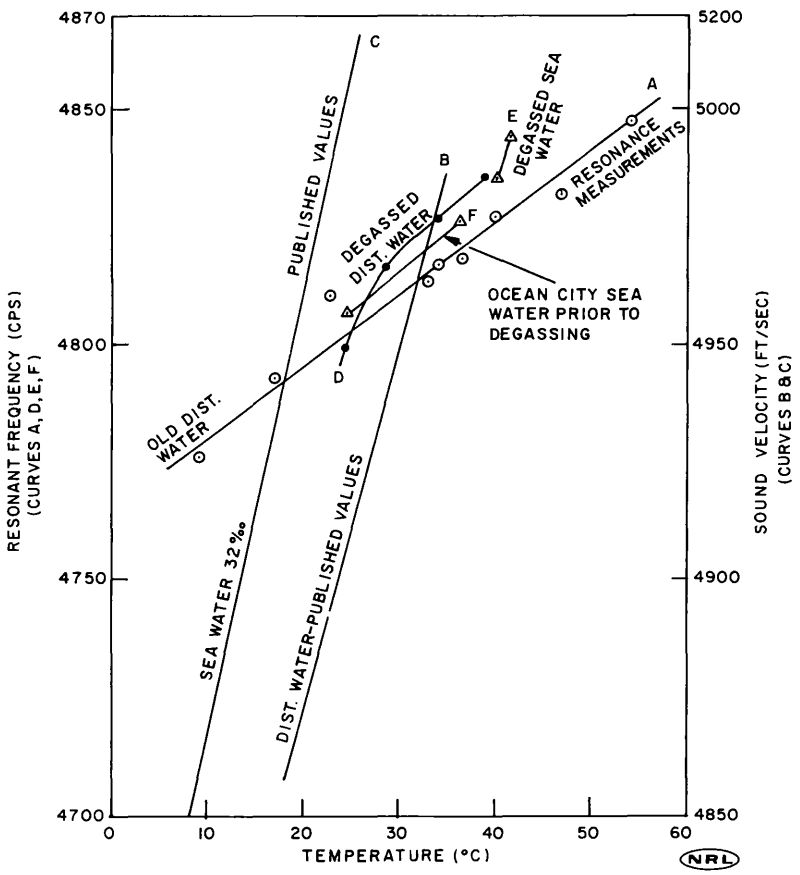

Fig. 2 - Comparison of acoustic resonance measurements with published values of sound velocity. The temperature changes were obtained by the immersion of the flask in a water bath.

Particles may play a major role in supporting free gas by serving not only as cavitation nuclei but as gas adsorbers. For a given mass of particles the surface-to-volume ratio of the particles becomes enormous as particle radii decrease. This vast area may well adsorb dissolved gas, resulting in particles having an air shell. The author is presently testing this hypothesis by resonance measurements on samples where particles are created in the liquid through chemical reaction. An example is the formation of a silver chloride precipitate formed in saline solution using silver nitrate. The addition of powdered material to the sample should not be utilized experimentally, since air may already be adsorbed by the powder.

CONCLUSIONS

1. Standing sea water and aged distilled water contain a stable bubble population as determined by acousitc resonance measurements.

2. Degassing of the water by vacuum-impact results in sound velocity-temperature data having the same slope as published data. The presence of bubbles decreases the slope.

RECOMMENDATIONS

1. A clear understanding of the nature of stable bubbles and the role of particles in bubble structure will require additional investigation, which should be pursued in the laboratory and in natural waters.
2. Additional research should be conducted on "ideal" water having no particles or dissolved foreign substance. This will require elaborate facilities to produce the water and conduct measurements in a contaminant-free environment.
3. An independent method for determining the presence of free gas should be established to verify the results and conclusions obtained by resonance measurements. A possible technique may be the measurement of compressibility, if sample-container reactions can be eliminated.

REFERENCES

1. Hiller, A.J., Mathes, R.H., and Ricalzone, L.C., "Ocean Surface Effects," Report of NRL Progress Sept. 1965, p. 37.

2. Harvey, N., et al., "Bubble Formation in Animals," Cellular and Comparative Physiology, 24:1 (1944)

3. Fox. F.E., and Herzfeld, K.F., "Gas Bubbles with Organic Skin as Cavitation Nuclei," J. Acoust. Soc. Am. 26:984 (1954)

4. Turner, W.R., "Physics of Microbubbles," Vitro Corp., Silver Spring, Md., Tech. Note 01654.01-2, 30 Aug. 1963

5. Turner, W.R., "Microbubble Persistence in Fresh Water," J. Acoust. Soc. Am. 33:1223 (1961)

6. Liebermann, L., "Air Bubbles in Water," J. Appl. Phys. 28:205 (1957)

7. "Acoustic Microbubble Spectrum Analyzer," Proposal of Vitro Lab. Silver Spring Md., SSL-C-12754 N-04335.94038, 7 July 1965

8. Hueter, T.F., and Bolt, R.H., "Sonics," New York:Wiley, 1965, p. 368

9. "The Physics of Sound in the Sea," Summary Technical Report of Division 6, NDRC, Vol. 8, p. 473, eq. 81

10. Del Grosso, V.A., "The Velocity of Sound in Sea Water at Zero Depth," NRL Report 4002, Jun. 11, 1952 p. 13, Fig. 1

11. Del Grosso, V.A., "The Velocity of Sound in Sea Water at Zero Depth," NRL Report 4002, Jun. 11, 1952, p. 16, Fig. 1-c

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY *(Corporate author)* Naval Research Laboratory Washington, D. C. 20390	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP

3. REPORT TITLE

SOME EXPERIMENTS WITH A LIQUID FILLED ACOUSTIC RESONATOR SHOWING THE EXISTENCE OF STABLE BUBBLES IN WATER

4. DESCRIPTIVE NOTES *(Type of report and inclusive dates)*

An interim report on the problem; work is continuing.

5. AUTHOR(S) *(First name, middle initial, last name)*

Alexander J. Hiller

6. REPORT DATE July 18, 1967	7a. TOTAL NO. OF PAGES 9	7b. NO. OF REFS 11
8a. CONTRACT OR GRANT NO. NRL Problem S01-26 b. PROJECT NO. SR 104-03-01-8136 c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) NRL Report 6567 9b. OTHER REPORT NO(S) *(Any other numbers that may be assigned this report)*	

10. DISTRIBUTION STATEMENT

Distribution of this document is unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Dept. of the Navy (Naval Ship Systems Command) Washington, D. C. 20360

13. ABSTRACT

Experiments were performed with a water-filled acoustic resonator to measure sound velocity in water as a function of temperature and undissolved gas. The measurements on gas-free distilled water agree well with theory, but measurements on sea water do not. This discrepancy can be accounted for by the existence of a stable microbubble population in the sea water, and by changes to this population as a result of the addition of thermal energy. Results showing that vacuum degassing can increase the sound velocity in the sea water sample demonstrated that a stable bubble population was present. Additional studies are needed for a clearer understanding of the nature of the stable bubbles and the role of particles in stable bubble formation.

DD FORM 1 NOV 65 1473 (PAGE 1)

S/N 0101-807-6801

Security Classification

Security Classification

14. KEY WORDS	LINK A ROLE	LINK A WT	LINK B ROLE	LINK B WT	LINK C ROLE	LINK C WT
Acoustic Scattering						
Acoustic Resonator						
Sound Velocity						
Microbubbles						
Vacuum Degassing						

DD FORM 1 NOV 65 1473 (BACK)
(PAGE 2)

GPO 927-606

Security Classification