NRL Report 6545

The Influence of Surface-Active Material on the Properties of Air Bubbles at the Air/Sea Interface

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May 8, 1967

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Washington, D.C.

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ABSTRACT

The lifetimes of single air bubbles were measured at water surfaces covered by various monomolecular films. Simultaneous measurements of bubble lifetime vs molecular area and film pressure vs area were made using air bubbles of various diameters. As the film pressure increased, the bubble lifetime passed through a maximum at about 1 dyne/cm and decreased at higher pressure with closer molecular packing. Mechanisms to account for this effect were based on surface tension gradient flow and dilational viscous forces within the film. The bubble stability data were used to describe molecular interactions and orientations within the stabilizing monolayer.

The lifetimes of bubbles at an air/sea interface were shown to be dependent upon the concentration of organic material in the water and the surface-chemical condition of the sea surface. The presence of a coherent surface film substantially reduced bubble stability, especially at elevated film pressures.

PROBLEM STATUS

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

AUTHORIZATION

NRL Problem H01-17
Project RR 002-10-45-5052

Manuscript submitted February 10, 1967.

THE INFLUENCE OF SURFACE-ACTIVE MATERIAL ON THE PROPERTIES OF AIR BUBBLES AT THE AIR/SEA INTERFACE

INTRODUCTION

Free gas bubbles in the sea may be produced by the decomposition of detrital matter, the impact of raindrops, and the breaking of waves. By scavenging surface-active material and foam-stabilizing particulate substances these bubbles promote the formation of sea foams, which under special conditions have been known to accumulate in large quantities in windrows and at the sea shore (1,2). Furthermore, the bursting of these entrained air bubbles at the sea surface may be an important source of the "giant" salt nuclei thought to be instrumental in the mechanics of maritime precipitation (3). According to Blanchard (4) a bursting air bubble generates salt-containing aerosols through the disintegration of the thin film of water which separates the air in the bubble from the atmosphere. Collapse of the bubble walls following the initial rupture of the thin water film results in the propulsion of several "jet" drops several centimeters into the atmosphere. These drops are about 100 microns in diameter, and their maximum ejection height is primarily a function of bubble size.

The presence of organic surface films may modify many processes occurring at the air/sea interface. Dramatic increases in capillary-wave damping (5), alterations of the sea surface temperature, and the inhibition of surface convectional cells (6) by monomolecular surface films have been demonstrated both in the laboratory and at sea. Surface-active substances, adsorbed at the air/sea interface as well as in the bulk sea water, also modify the stability and bursting properties of air bubbles. It has been demonstrated qualitatively (4) that the natural surface-active compounds of the sea are transferred into the marine atmosphere along with the sea water droplets produced by the bursting bubbles. Once the droplet water has evaporated, the remaining salt residue becomes partially coated with an adsorbed film of surface-active molecules. If this occurs, the wettability and consequently the cloud-droplet nucleating efficiency of the salt particle will be decreased, since the particle surface will have a nonzero contact angle to water. According to Fletcher (7) much higher cloud supersaturations are required for the nucleation of a water droplet by a nonwettable condensation nucleus.

One of the peculiarities of air bubbles at the air/water interface has been observed by Blanchard (4) and Day (8), who found that in some instances air bubbles did not burst instantaneously when they reached the water surface. They attributed this bubble stability of variable duration to contamination of the water. This stabilization of air bubbles by surface films was first examined in detail by Hardy (9). Lifetimes of single air bubbles at a water surface containing a monomolecular layer of oleic acid were measured as a function of the surface tension. No stability was imparted to the bubble by the monolayer until the molecules were sufficiently compressed to decrease the surface tension. A maximum lifetime was attained at a film pressure (surface tension decrease) of about 1 dyne/cm, and with further increases in the film pressure (F) the lifetimes decreased to near zero at high values of F. This unusual and unexpected effect was examined in more detail by Trapeznikov (10) and Brown et al. (11). A number of fatty acids and esters were examined, all of which gave similar results in that the bubble lifetimes passed through a maximum value at low F and approached zero at the collapse pressure of the monolayer (high F).

To understand better the generation and persistence of sea foams as well as the properties of air bubbles at the sea surface the present study was directed toward the effect of known monomolecular films on the stability of single air bubbles. Bubble lifetimes at water surfaces were studied as a function of film pressure, molecular orientation, and dynamic surface properties of the stabilizing monolayer. Fresh sea water samples were included in this work to relate the controlled laboratory data to the natural ocean environment.

APPARATUS AND METHODS

Bubbles of various diameters (0.7 to 5.1 mm) were produced from hypodermic needle tips, Teflon capillary tubing, and drawn glass tubing. Single bubbles, generated at a controlled rate and size by a Harvard syringe pump operated at 0.101 ml/min, were ejected 1 cm below the surface of triply distilled water contained in a hydrophil tray. The sides of the tray were coated with a low-surface-energy perfluoroester resin, and waxed glass sliding barriers were used to clean the water surface and to compress applied monomolecular layers. Pure monolayer-forming compounds were added to the water surface from dilute chloroform or hexane solutions with a calibrated microsyringe. Surface tensions were measured by the ring method with a du Nouy tensiometer. The term film pressure, which will be used frequently, is the difference between the surface tension of clean water and that of a monolayer-covered surface. Experiments were performed in a temperature-controlled room at 25 ± 0.5°C.

To obtain reliable surface-chemical data, high reagent purity and quantitative procedures are essential. Polar contamination was minimized through the use of triply distilled water, the final two stages of distillation being from a quartz still. The water surface is swept a number of times with the waxed barrier prior to the introduction of monolayer-forming compounds. These precautions were necessary since any soluble polar impurities would adsorb at the bubble/water interface as well as at the water surface and lead to increased bubble stability. Fatty acid monolayers were spread on 0.01-N sulfuric acid substrates to avoid heavy metal soap formation from contaminant ions. Small amounts of heavy metal salts would drastically alter the surface properties of the fatty acid monolayers (12).

Synthetic sea water substrates were not studied because of the almost impossible task of attaining freedom from water-soluble organic contaminants adsorbed on the salt constituents. For this reason, only distilled water and acid substrates were studied to demonstrate the fundamentals which govern bubble stability. These concepts will then be used in conjunction with data from natural sea water experiments to explain bubble properties in the marine environment.

EXPERIMENTAL RESULTS

Bubble lifetimes (T) and film pressures (F) are plotted simultaneously against the area per molecule (A) for a number of pure monolayer-forming compounds (Fig. 1). The selected pure compounds provided a variety in chemical structure and monolayer properties. Some of the compounds gave rigid close-packed monolayers upon compression, while others were expanded films which behaved like two-dimensional gases and liquids. There were also regions of high compressibility (low dF/dA) represented by plateaus in the F vs A curves (Figs. 1a, 1e, and 1f) for several of the monolayers.

In all cases the bubbles burst spontaneously upon reaching a clean water surface, as long as no surface-active material was present in the bulk water. That is, once the air/water interface of the bubble met the water surface, rupture was almost immediate when no polar organic materials were present in either interface to stabilize it against rupture.

It should be mentioned that any soluble surface-active material in the bubbled water can adsorb at either or both air/water interfaces and increase the bubble lifetime. Such contamination can modify the results significantly and should be eliminated by careful cleaning of equipment and purification of reagents.

As the relatively water-insoluble surface films were compacted by the sliding barrier, and the surface concentration was increased, no bubble stability resulted until there was a measurable film pressure (Fig. 1). With further surface compression the bubble lifetimes (T) increased rapidly to a maximum value at low F (0.5 to 1.5 dynes/cm) and decreased at higher film pressures. Once the monolayer had collapsed, T values were usually 0 to 2 sec. One of the siloxane monolayers had a region of high compressibility or plateau in its F vs A curves (Fig. 1a). T values remained constant in this compressible region even though A was decreasing. Once these monolayers were compressed to a point where compressibility (dF/dA) decreased, the bubble lifetime decreased and the molecules of this monolayer became close-packed. T values for several monolayers decreased steadily with decreasing A after the maximum value had been reached. On the other hand, T did not decrease to any great extent until the monolayer had collapsed in other experiments (Figs. 1c, 1d). These data will be interpreted in the ensuing discussion.

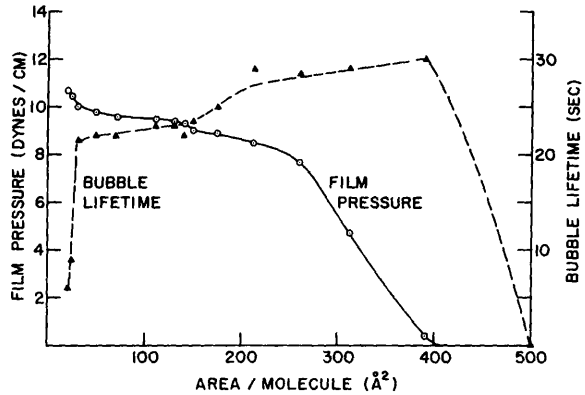
The effect of bubble diameter on the bubble stability is listed for several experimental situations (Table 1), and T vs A is plotted for two bubble diameters for an oleic acid monolayer (Fig. 1b). T increased with bubble diameter in a nonlinear fashion up to diameters of about 4 mm. Bubbles of greater size were unstable and irreproducible. In Fig. 2 plots of T vs A for two bubble diameters show that T for the 3.2-mm bubble was greater than that of the 2.0-mm bubble throughout the compression of the oleic acid monolayer. A similar experiment using a 4.2-mm bubble produced a T vs A plot which coincided with that for the 3.2-mm bubble. 2.0-mm-diameter bubbles were used for all of the data reported unless otherwise stated.

In most of the experiments the precision of the T values was sufficiently high that the measurement of bubble lifetime may be considered to be a dynamic physical property of the system. For example, the average deviation of T at ten different film pressures for an oleic acid monolayer using a 2.0-mm bubble varied from ±0.2 to ±0.5 sec. When compared to the magnitude of T the percent average deviation varied from 5.4 to 7.5. In general the precision of most of these data fell within ±8%. If precautions are not taken to eliminate soluble surface-active contamination, T values vary widely. In addition, at high F it was necessary to wait about one minute between measurements to allow the monolayer to reach equilibrium in the area affected by a previous bubble. Average deviations as high as ±60% resulted when successive measurements were made without such a waiting period.

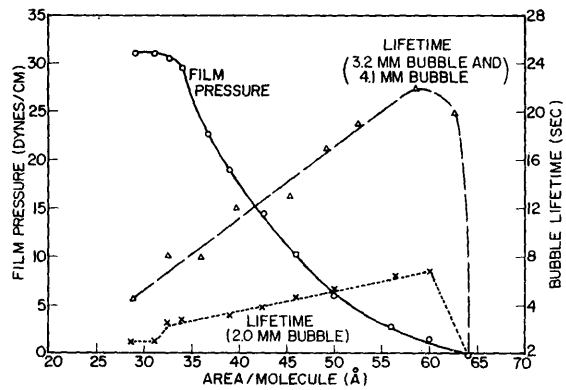
DISCUSSION

Some information about the molecular processes occurring within a monolayer as it is compressed may be derived from F vs A data. While the slope (dF/dA) of the force-area curve is a measure of the two-dimensional state of the film, the surface area per molecule at which changes in slope occur is often indicative and descriptive of reorientations of the surface molecules. Additional information on surface conformation may be derived from capillary-wave-damping data (13,14). The present study of bubble lifetimes as a function of F and A for a number of chemically different monolayers will be considered as another dynamic method for probing surface processes.

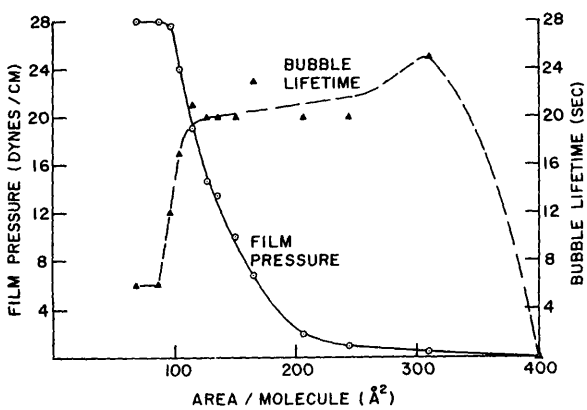
In the ensuing discussion of monolayer-produced bubble stability reference will be made to Fig. 2, a schematic of a single air bubble at a monolayer-covered surface. By definition the thin layer of water between the two air/water interfaces will be called the



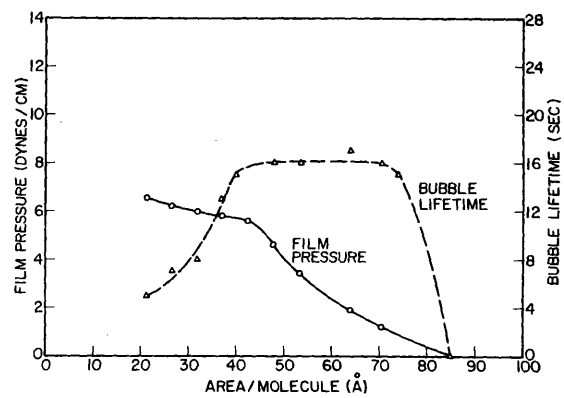
(a) Polydimethylsiloxane (dodecamer) monolayer on distilled water



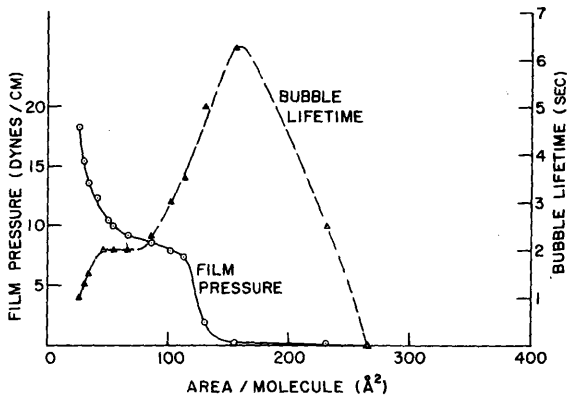
(b) Oleic acid monolayer for 3.2 and 2.0-mm-dia. bubbles on an acid substrate, pH = 2



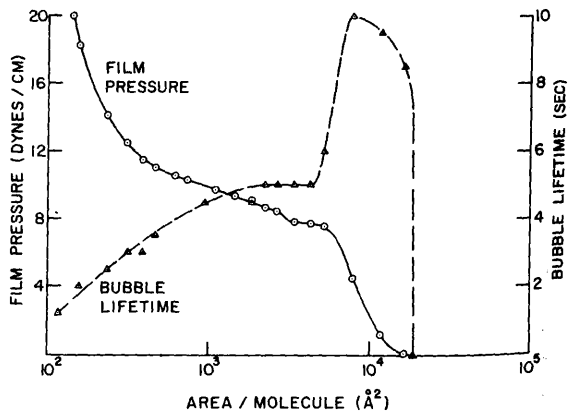
(c) Polydimethylsiloxane (heptamer) monolayer on distilled water



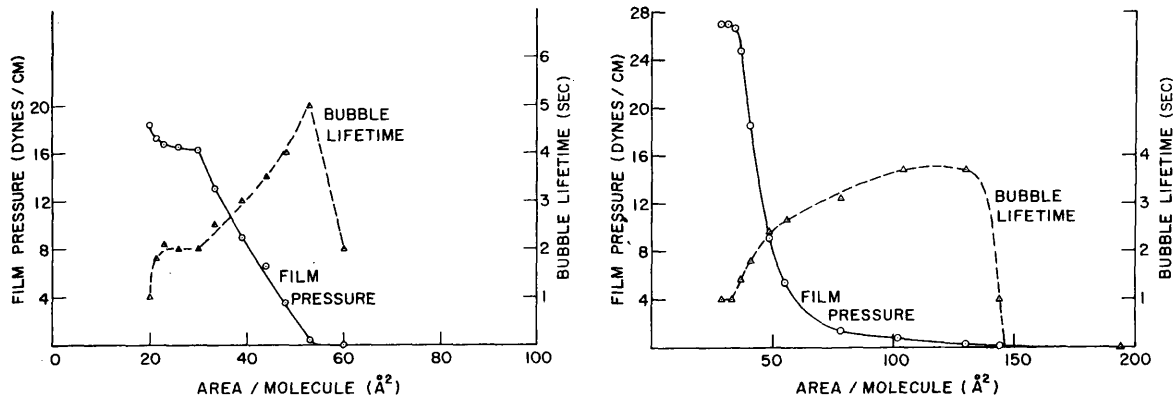
(d) Amyl stearate monolayer on distilled water



(e) 12-hydroxystearic acid monolayer on an acid substrate, pH = 2



(f) Polyethylene glycol monolayer on distilled water



(g) Tetradecanoic acid monolayer on acid water, pH = 2

(h) Linolenic acid monolayer on acid water, pH = 2

Fig. 1 - Dependence of bubble lifetime upon the area per molecule of a surface monolayer (dashed line) and corresponding film pressure vs molecular area curve (solid line)

Table 1
Influence of Bubble Diameter on Bubble Lifetime

Bubble Diameter (mm)	Bubble Lifetime (sec)		
	Oleic Acid		12-Hydroxystearic Acid
	F=12.2 dyne/cm	F=2.0 dyne/cm	F=0.5 dyne/cm
0.70	-	1.1	-
1.0	1.5	2.2	1.0
1.5	-	-	3.9
2.0	4.0	5.1	7.0
2.5	7.2	10.3	-
3.2	12.1	21.0	14.5
4.1	-	22.1	14.0

bubble film, the raised portion of the bubble above the still water surface, is the bubble crown, and the spread insoluble monomolecular layer will be referred to as a monolayer or surface film. The infinitesimal (-dF) represents a temporary decrease in film pressure created by a local increase in surface area above the bubble crown. Should the surface film possess solubility, the shortest path to the bubble/water interface is at the thinnest portion of the bubble film (solubility path). Processes important to the stability of the bubble are indicated on the schematic.

The bubble-stability maximum and its subsequent decrease with increasing F can be explained in terms of two opposing surface effects. As film-forming molecules are added to the surface, T values are zero until there is a potential surface-tension gradient ($\Delta\gamma$) of sufficient magnitude to drive protective molecules into the expanded surface area created by the protruding bubble crown. In this region molecules are further apart and

the surface tension is greater. The surface tension gradient flow is in opposition to the drainage of water in the underlying bubble film. The decrease in T which occurs at greater film pressures can be attributed to an increase in intermolecular cohesion between molecules in the protective film. Molecular interactions increase as the molecules are forced into closer contact and become adlineated. This hysteresis effect resists the gradient flow into the area of greater surface tension created by the bubble crown. As cohesive forces increase between molecules, changes in surface tension above the bubble are not immediately counterbalanced. Thus, potential holes or points of rupture exist in the protective surface film, and T is decreased.

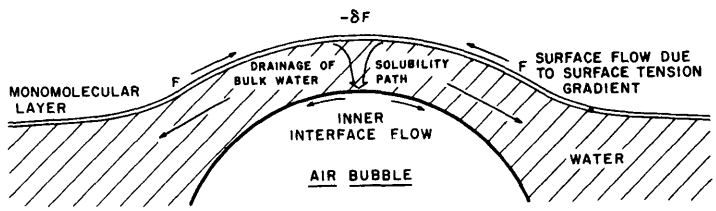


Fig. 2 - Factors affecting the lifetime of an air bubble at a water surface

These conclusions are substantiated by the data presented in Fig. 1. When the concentration of surface molecules was low, dF/dA was low, and no stability was imparted to the air bubble. T increased rapidly to a maximum value when the monolayer entered a more condensed state and the surface tension gradient increased. Bubble lifetimes decreased when the surface molecules became close-packed, and voids in the surface film were not repaired even though dF/dA remained high.

Information concerning the configuration of molecules within the spread monolayers can be obtained from these data. Several of the monolayers–amylstearate, 12-hydroxystearic acid, the polyorganosiloxanes, and the polyether (Figs. 1a, 1c-1f)–had portions of their T vs A plots in which T was approximately constant and did not decrease with decreasing area per molecules. These plateaus in some cases coincided with compressible portions of the corresponding F vs A curves. Thus the decrease in A was not placing molecular chains in sufficiently close contact to cause an increase in cohesional forces, although certain molecular rearrangements were occurring. For example, force-area data by Fox et al. (15) for polydimethylsiloxanes revealed a plateau region of high compressibility. It was postulated that the siloxane molecule coiled into a helical configuration such that hydrophobic methyl groups were on the outside of the coil. The long axis of the coiled molecule was parallel to the water surface with the methyl groups as nearest neighbors to the water substrate. Wave-damping data by Garrett and Zisman (13) showed low monolayer-substrate interactions at this plateau region of the F vs A curve, further substantiating the coiled model. From Fig. 1 we see that bubble stability does not decrease markedly for the siloxane monolayer until the film becomes incompressible at low molecular areas and the helical coils have been forced into an upright, adlineated orientation. The bubble stability data are also consistent with the model for the siloxane coil. The process of coiling does not greatly increase intermolecular cohesions as A is decreased, and surface flow driven by surface tension gradients is consequently unimpeded.

A possible interpretation of the results for amyl stearate (Fig. 1d) is that the amyl group must be lifted upon compression along with the longer stearyl chain. Close-packing of the longer chains is prevented by the amyl groups, and T does not decrease until the monolayer has collapsed at about 40 $Å^2$/molecule (an area corresponding to two upright adlineated hydrocarbon chains per molecule). For amyl stearate as well as for many

other molecules T decreases to near zero after collapse of the monolayer. This is a likely result of strong interactions between the molecules of a randomly oriented multilayer. Monolayer collapse is often clearly marked by abrupt decreases in T.

The polyethylene glycol monolayer (Fig. 1f) is difficult to interpret because of the indeterminate composition of the commercial preparation. Not only is the polymer a mixture of molecular weight sizes, but the chemical nature of the end groups is not known. In spite of this several interesting observations can be made. Unlike the siloxane data, T for polyether decreased with A in two stages. The initial T decrease could be attributed simply to a decrease in dF/dA as shown by the F vs A plot rather than by a specific molecular rearrangement. Further decreases in T may be due to a gradual collapse of the monolayer or to an adlineation of polymer molecules lifted from the surface.

Figures 1b, 1g, and 1h are data for molecules which lift gradually from the water surface as they are compressed without special coiling or other rearrangement. Consequently, T decreased steadily with decreasing A after the maximum had been passed. Compounds which gave condensed films (cetyl alcohol and 9,10-dihydroxystearic acid–not plotted) gave low maximum values of T of from 2 to 3 sec. For these compounds close molecular packing already existed by the time the bubble-stabilizing surface tension gradient had been established, and the surface film did not flow readily into the expanded area of the bubble crown.

The mechanisms proposed for the stability of single bubbles beneath an insoluble monolayer are also useful in explaining the variability of data at high and low F. At low F, in the neighborhood of the minimum surface tension gradient necessary for stability, depletion of the surface film by the bursting bubble temporarily reduces the local surface concentration. If bubbles are to be generated at the same point, time must be allowed for the surface film to restore itself to the original condition. At high film pressures, time must also be allowed between measurements to obtain repeatable values of T. In this instance hysteresis effects prevent the monolayer from rapidly filling the gap left by the bursting bubble. For example, T values taken at equilibrium surface condition might be 2 sec at a high F. If taken in rapid succession the bubbles released beneath the same monolayer would burst almost instantaneously upon reaching the surface.

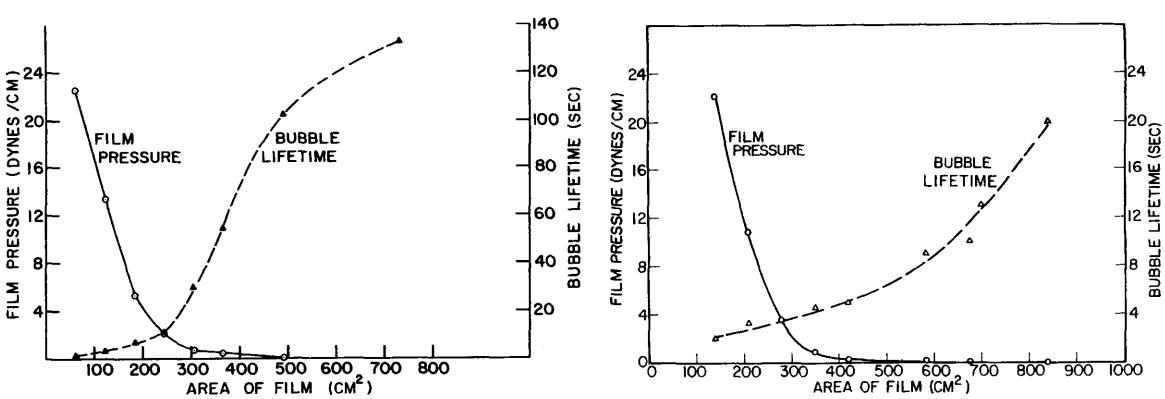
To understand the increase in T with bubble diameter it is necessary to consider the shapes of air bubbles at the air/water interface. Toba (16) found that bubbles up to about 2-mm in diameter were roughly spherical in shape, whereas bubbles of greater size became increasingly more flattened (greater breadth than depth) and protruded further above the still water surface. As a consequence of this change of shape, the area of the bubble crown which protrudes above the water surface increases rapidly with increasing bubble diameter. Attempts were made to correlate the lifetimes of bubbles of various diameters (Table 1) with corresponding bubble crown areas calculated from Toba's data. Although both factors increased with bubble diameter, no simple relationships were apparent. However, the results may be explained qualitatively in light of the expanded surface area created by the protruding bubble. The small bubbles with little increase of surface area caused only small local dilution of the molecules of the stabilizing monolayer. Larger-diameter bubbles, on the other hand, caused greater separation of the surface molecules. Thus, surface flow under the driving force of the surface tension gradient was less impeded by attractive forces between molecules for the larger bubbles, and T was greater. When the increase in the area of the bubble crown became very large (diameter > 4 mm), T values became unreliable, since the surface concentration was insufficient to cover the bubble film completely and protect it against rupture.

BUBBLE STABILITY IN SEA WATER

Bubble lifetimes were measured on freshly collected sea water using the 1-cm-deep hydrophil tray and 2.0-mm bubbles. The tray was filled, the water surface was swept clean with the sliding hydrophobic barriers, and T values were measured at intervals during a 1-hour period. The surface-active material which had reached the water surface during the 1-hour aging period was then compressed in stages, and T and F were measured at each film area.

During the initial aging period the bubbles were stabilized by soluble and insoluble surface-active material which had been scavenged by the bubble on its rise to the surface. This film was adsorbed at the air bubble/water interface, thereby stabilizing the bubble film against immediate rupture at the surface. In organically rich sea water the T values were somewhat greater than those caused by the previously described insoluble monolayers. This greater stability may be due to the rapid repair of the protective surface film by the available supply of adsorbable material from the bulk sea water. As might be expected, local concentration gradients caused by the passage of previous bubbles resulted in large variations in the measured bubble lifetimes. For example, sea water sample A, collected 5 miles south of Cape Charles, Virginia, had values which varied from 30 sec to several minutes.

Once the surface had aged and the adsorbed surface film was compressed, T decreased substantially (Fig. 3a) and the repeatability of the measurements increased. As F increased due to compression of the surface film, the magnitude of T approached that for insoluble monolayers. Thus, although surface-active material is present on the inner air bubble surface, the monolayer at the atmosphere/sea water interface was the controlling factor in determining T.



(a) Sea water sample A

(b) Sea water sample B

Fig. 3 - Bubble lifetime (dashed line) and film pressure (solid line) vs area of surface film of sea water

Sample B was a near-surface collection from less fertile water of the Florida Current, 4 miles east of Ft. Lauderdale, Florida. T values were much less during the aging period than for the more fertile sample A. Some values of T were zero, since previous bubbles had removed surface-active material from the vicinity of the bubble generator. Moving the generator to another location usually gave stable bubbles. In other instances several zero values were obtained before sufficient material had been carried into the water surface to stabilize a subsequent bubble. It should be noted at this point that a gas bubble is an important agent for the transport of organic surface material to the sea surface (5). However, when a bubble bursts some of the accumulated surface film is removed and is carried into the atmosphere by the escaping jet drops (4). The

bubble, therefore, is responsible for a net movement of surface-active material from the sea, across the air/sea interface, and into the marine atmosphere.

From these data it is possible to predict some of the effects that natural and/or artificial monolayers (sea slicks) would have on oceanic processes. A well-compressed sea slick would reduce bubble lifetime and consequently act as an antifoaming device. That is, once the slick molecules are compressed so that the film behaves like a two-dimensional liquid or solid, the bubble lifetime is determined largely by the surface film rather than the adsorbed film picked up by the bubble on its rise to the sea surface. In the absence of a surface slick, bubbles at the sea surface would have long lifetimes if they had been exposed to chemically fertile water during their rise through the ocean.

CONCLUSIONS

The lifetime of a single gas bubble at an air/water interface beneath an insoluble monolayer is determined by the physical properties of the surface film. No stability is imparted to the bubble by the adsorbed surface film until the molecules are in close enough contact to generate a film pressure. A surface tension gradient is then established when surface dilations occur. This gradient drives the protective monolayer over the bubble crown against the counter flow of the draining water in the bubble film. Bubble lifetime reaches a maximum at about 1 dyne/cm film pressure near the transition point of the monolayer from a gaseous or highly compressible state to one of less compressibility. The decrease of the bubble lifetime from the maximum value as the monolayer is further compressed is attributed to a rigidity or hysteresis of the monolayer due to increased intermolecular cohesional forces. This resistance to dilational flow opposes the surface gradient flow in determining the resultant bubble stability.

Bubble stability can provide useful qualitative information about molecular interactions and orientations within the stabilizing film. When used in conjunction with film pressure and capillary-wave damping measurements, a picture of molecular configurations and monolayer-substrate interactions at a liquid surface can be developed.

The lifetime of bubbles at an air/sea interface is a function of the fertility of the water and the surface-chemical condition of the sea water surface. In the absence of a surface film, bubble lifetime is determined by the surface-active material adsorbed onto the bubble during its passage upward through the bulk water. In general this stability is much greater than that induced by a surface film. The presence of a monomolecular layer at the air/sea interface will substantially reduce bubble stability even though the underlying water may be rich in surface-active organic substances. The greater the pressure on the surface film, the less will be the lifetime of an individual bubble.

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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

THE INFLUENCE OF SURFACE-ACTIVE MATERIAL ON THE PROPERTIES OF AIR BUBBLES AT THE AIR/SEA INTERFACE

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

An interim report on one phase of the problem.

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

W.D. Garrett

6. REPORT DATE

May 8, 1967

7a. TOTAL NO. OF PAGES

14

7b. NO. OF REFS

16

8a. CONTRACT OR GRANT NO.

NRL Problem H01-17

b. PROJECT NO.

RR 002-10-45-5052

c.

d.

9a. ORIGINATOR'S REPORT NUMBER(S)

NRL Report 6545

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

Department of the Navy (Office of Naval Research) Washington, D.C. 20360

13. ABSTRACT

The lifetimes of single air bubbles were measured at water surfaces covered by various monomolecular films. Simultaneous measurements of bubble lifetime vs molecular area and film pressure vs area were made using air bubbles of various diameters. As the film pressure increased, the bubble lifetime passed through a maximum at about 1 dyne/cm and decreased at higher pressure with closer molecular packing. Mechanisms to account for this effect were based on surface tension gradient flow and dilational viscous forces within the film. The bubble stability data were used to describe molecular interactions and orientations within the stabilizing monolayer.

The lifetimes of bubbles at an air/sea interface were shown to be dependent upon the concentration of organic material in the water and the surface-chemical condition of the sea surface. The presence of a coherent surface film substantially reduced bubble stability, especially at elevated film pressures.

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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
Surface-active material						
Air bubbles						
Air/sea interface						
Monomolecular films						
Oceanography						
Sea surface chemistry						
Foam stability						

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Security Classification