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**A Review of Experimental Observations and Remaining
Questions Concerning Formation, Persistence,
and Disappearance of Sea Slicks**

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13. ABSTRACT (Maximum 200 words) Over 500 articles related to questions concerned with understanding the forces that act to control the formation, persistence, and disappearance of sea slicks and to determine their properties have been gathered and cross-indexed under a set of key words. A brief review of current thinking about slicks drawing on the database of listed references is presented. Recent results from the literature comparing dynamic and static surface dilational modulus data are summarized. To provide an illustrative example for the general discussion, new data for a single natural film formed on the surface of a seawater sample are presented. The natural sample was characterized by determining its film pressure, modulus, surface potential, and corresponding damping coefficients for 30 Hz capillary waves as a function of the area occupied by the film. These were compared to corresponding data for two pure compounds that form monomolecular films on water. Previously unpublished data on the spontaneous spreading rates of 10 pure surface-active compounds were used to determine the relationship between spreading velocity and surface tension gradient. Topics in need of further study where data for natural films are scarce or nonexistent are identified.			
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A REVIEW OF EXPERIMENTAL OBSERVATIONS AND REMAINING QUESTIONS CONCERNING FORMATION, PERSISTENCE, AND DISAPPEARANCE OF SEA SLICKS

BACKGROUND

Introduction

This report has been prepared to gather together information from the literature and from a small additional quantity of previously unpublished data relating to the formation, behavior, persistence, and disappearance of sea slicks on the surface of the ocean. A particular interest exists in attempting to better understand the behavior of slicks associated with the passage of ships through the water. Over the years, several recurring questions about slicks have arisen, and not all of the questions have been fully answered to the satisfaction of everyone concerned. Some of these questions are (1) Which physical properties of the air/water interface are altered by the presence of surface-active organic compounds? (2) How do these properties change with a change in the concentration of surfactant per unit area? (3) What is the source of the material concentrated at the surface? (4) What are the transport mechanisms and their relative rates? (5) How are properties of slicks related to wind speeds and surface current velocities? and (6) Why do slicks persist for such a long time after they are formed?

Although these questions will not be answered definitively in this report, an attempt has been made to gather much of the information related to these questions from the literature to provide insight into the probable answers. More than 500 articles and reports containing information related to slicks have been identified and gathered into the list of references at the end of this report. Only a brief synopsis of what is known about slicks is given in the text, and an attempt is made to point the reader to the evidence in the literature that supports the assertions.

Although a large number of references have been identified, this review undoubtedly has overlooked many others. A deliberate attempt was made to include older literature, government reports, and contract research reports that are not always uncovered in modern library computer database searches. Many of these items were never entered in computer databases because they predate the existence of such modern library tools. Although much space is required, references have been given as completely as possible to make retrieval of the information easier.

The report begins with a very brief overview of the characteristics of slicks and factors that influence their formation and dissipation. Immediately after the overview, we clarify the terminology often encountered when examining the multidisciplinary literature concerned with this subject. The assertions contained in the overview are later supported by brief summaries of the evidence that have led to them and by the identification of several additional sources of information contained in the list of references. A more extensive listing of the literature concerned with many of the more important topics is given by the key word cross-reference list that appears just before the references. Areas where additional experiments are required to confirm or modify currently held beliefs are identified in the course of exploring the evidence related to each topic.

Summary of Characteristics Attributed to Slicks

Slicks are regions of damped capillary waves on natural water bodies that can be seen by eye. They are visible because of the difference in the degree of light reflectance from the damped region relative to the background area. Radars operating at certain frequencies can also detect the areas of damped waves. Naturally occurring, surface-active organic molecules that adsorb at the air/water interface are believed to be primarily responsible for the wave-damping effect. Adsorbed organic compounds are distributed widely over the surface of natural water bodies. It is only when these polar organic compounds on the water surface are brought sufficiently close together to alter the physical properties of the interface, that slicks are observed. The naturally occurring, surface-tension-reducing compounds probably act together with other physical mechanisms to produce the damped regions.

Association With Water Motion – A slick can be produced by man on wave-rippled water that has no subsurface motion by the deliberate spreading of a surface-active organic film [20,27]. However, natural slicks may be associated with surface water convergences and motion of the underlying bulk water that produce the convergences. A persistent Langmuir circulation cell will contribute to the persistence of a slick. Naturally occurring periodic banded slicks observed in coastal waters near San Diego, California, have been shown to be associated with convergences caused by internal waves [137,138,298,299]. Slicks associated with the wake of ships passing through the water may be formed into organized patterns by surface water convergences induced by the ships.

Source and Physicochemical Properties of the Surface-Active Material – The source of the film-forming material is believed to be the biological organisms in the sea, especially the microorganisms such as phytoplankton and is generally not caused by pollution by man. Analysis of material associated with slicks has revealed a quite complex mixture of organic compounds (see references listed under the key word **analysis** in the cross index). A significant fraction of this material apparently has a rather large average molecular weight (> 2000) suggesting polymeric compounds, but the carbon-hydrogen-oxygen-nitrogen analysis suggests a higher concentration of carbohydrates than proteins. Every sample of seawater examined has contained a detectable quantity of surface-active material, including samples from as deep as 2000 meters [35]. Much higher concentrations of this material are usually observed near the sea surface.

Accumulation, Transport, and Loss Mechanisms – When samples of seawater collected even from non-slicked regions are allowed to stand, film concentrates at the surface and can be detected by measuring the reduction of surface tension. Detectable films form rapidly (in minutes) on seawater suddenly made still, and there is a slow increase in the quantity of material accumulating at the surface as time passes. Diffusion to and adsorption at the air/water interface of rising bubbles is a mechanism for scavenging surface-active material more rapidly from the bulk water and for increasing its rate of arrival at the sea surface. No striking difference appears to exist between surface-active material found at the sea surface and that found in bulk water with respect to the physical effects at the air/water interface [35]. Enriched concentrations of organisms and non-surface-active chemicals are often found associated with microlayer water collected from slick-covered areas. This is especially true in coastal waters and salt marshes. The use of oil-eating microbes has been advocated to clean up large oil spills, so it should not be surprising that one method of removal is consumption by feeding organisms. Wind-driven slicks have been observed to move at 3 to 5 % of the wind speed. Slicks can be driven by the wind onto the beach. Adsorption on suspended particles is another method of removal from the air/water interface. When the weather is violent, downward mixing into the bulk water and transport into the marine atmosphere by bursting bubbles associated with whitecaps probably represent the major loss mechanisms.

Reasons for Persistence of Slicks – The fact that persistent convergence of surface water can accumulate surface-active material and maintain a slick has been mentioned above. In the absence of water motion, forces act within a compressed film causing it to expand greatly at first, but also to remain coherent after expansion. A compressed monomolecular surface film may be conveniently thought of as a two-dimensional (2-D) gas under pressure. (Film pressure is defined as the difference in surface tension from clean water.) When the confining force or barrier is removed, a slick formed of surface-active material that is sufficiently concentrated to reduce surface tension will expand spontaneously until no gradient of surface tension exists. It has been observed in the laboratory that the maximum in the wave damping effect occurs at extremely low, and sometimes undetectable film pressures, so a greatly expanded film can still make a visible slick (i.e., intensity of damping is not a simple linear function of surface tension). Surface (electrical) potential measurements have shown that natural surface films contain oriented dipolar molecules. When the film is compressed, the dipoles stand vertically and like charges on the heads are repulsive (as are the like charges on the tails of the molecules). The film expands until this electrostatic repulsive force is balanced when the dipoles lie flat. Further expansion would have to overcome the coulombic attractive forces between the now unlike charges in the plane of the film, so in the absence of stronger dissipative forces the film remains coherent.

Terminology

Many individuals from various professional disciplines have studied the sea surface, and have occasionally interchanged terms appropriate to different fields even though the meaning is not exactly identical. Therefore, it is necessary to specifically define the meaning of some of the terms that are used throughout this report.

Slick – The term slick indicates a region of the surface of a natural body of water that is different in appearance from the surrounding water surface because the capillary waves and short gravity waves are sufficiently damped to produce an area that is visible to the eye and that may be seen in photographs or in images generated by certain radars. The fine-scale roughness is diminished. The phenomenon that is sensed by the eye is the relative depression of the very short waves. The layer of material that causes the damping is so thin (less than the wavelengths of visible light) that the material itself is not usually detected remotely. Sensors that operate in other wavelength regions besides that of visible light or radar may be able to detect the wave damping. By mentioning those particular sensors we do not mean to restrict the definition. Slicks may appear brighter or darker than the surrounding region. This often depends on the viewing angle. It is believed that most slicks observed on the ocean are the result of the presence of naturally occurring surface-active material and their film thicknesses are on the order of the dimensions of single molecules, i.e., less than 10 nm.

Oil Slick – An oil slick is caused when petroleum products are spilled on the sea. These pollutant slicks are man-made, their chemical composition is radically different from natural slicks, and they often are much thicker. Because of a density near that of water and cohesive forces in the oil, heavy fuel oil spilled on the sea can cover large areas with thicknesses measured in centimeters. Relatively thin oil slicks are often detected by observing a silvery sheen or interference colors. In these cases the oil itself is being observed rather than its effect on the capillary waves. Colored oil films are hundreds of nanometers thick [29]. Black oil films are much thicker. Pollutant oil films can damp capillary waves, and if they have become partly oxidized and spread thinly enough, the slicked area of water covered by them can be mistaken for a natural slick by the casual observer. However, spilled petroleum is rare compared to natural film-forming material, and films with high hydrocarbon content are easily detected by chemical analysis.

Monolayer; Monomolecular Layer – A monolayer is a single layer. In the context of air/water or air/solid interfaces, the term usually refers to a single layer of atoms, molecules, cells, or particles. The components do not have to be so closely packed as to be in contact with each other to still be referred to as a monolayer. The term "sub-monolayer" is often used to describe this condition of an expanded monolayer. A monomolecular film refers specifically to a film of chemical compounds that is only one molecule thick. Slightly soluble and insoluble surface-active organic compounds adsorb at an air/water interface to form monomolecular films. Most surface-active compounds are more attracted to water than to each other and therefore will not spread over their own monolayer. Therefore, single-molecule-thick films can easily be formed on the surface of water by spontaneous spreading of these substances from bulk material or from bulk material dissolved in a solvent. It is believed that naturally occurring surface-active material that can form monomolecular films at the air/sea interface and thereby reduce the attractive forces between the surface water molecules is primarily responsible for the regions of damped capillary waves called slicks.

Surface-active – The term surface-active in this report refers to the property of polar organic compounds that causes them to concentrate at an air/water interface. In general, a surface-active organic compound contains oxygen with unpaired electrons or some similar hydrophilic group at one end of the molecule and a relatively insoluble hydrophobic segment such as a straight hydrocarbon chain at the other end. The water-loving end of the molecule could dissolve, but the hydrophobic part prevents complete solubility, hence the molecules form a single layer at an air/water interface. Surface-active compounds can be soluble. However, at equilibrium in still systems they concentrate at interfaces. The presence of such materials on a water surface is most often detected by measuring the surface tension. When these materials are sufficiently concentrated at the water surface, surface tension is reduced. A partial charge separation exists between the two ends of surface active molecules giving them a polar character. Another method of detection is to measure the change in surface potential at the interface caused by increasing the density of oriented dipoles when films of these materials are compressed into smaller areas. Once a surface-active compound has reached the air/water interface, a considerable amount of energy is required to force it back into solution.

Surfactant – Surfactant is a short form of the term surface-active agent. This term is primarily used to describe synthetic industrial products and has a connotation implying that the substance is man-made. A detergent is a specific type of surfactant. Surfactants can form monomolecular films by adsorption at an air/water interface. Depending on the audience, it is often necessary to use the phrase natural surfactants to describe the material found in seawater to avoid confusion with man-made pollutants.

Surface Tension; Film Pressure – The surface tension of a liquid is a force that acts to minimize the surface area, because the attractive forces acting on the very surface layer of molecules are not balanced. Molecules within the bulk liquid are surrounded on all sides by other molecules that are attracted to them by cohesive forces. Because no corresponding molecule is present on the air side of an air/water interface, the surface molecules are attracted inward. Because the result is a minimization of surface area, surface tension forces are treated as forces that act parallel to the plane of the surface along a line. Hence, the units are usually expressed as dynes/cm or milliNewtons/meter. The presence of a single-molecule-thick layer of different molecules on the surface of water alters the balance of forces and reduces the surface tension. Film pressure is defined as the difference between the surface tension of clean water and the surface tension of water covered by a film. Changes in capillary wave velocity and wave damping are associated with changes in surface tension.

Units of Surface Tension or Film Pressure – In the cgs system of units surface tension and film pressure are reported in dynes/cm, a force per unit length. The numerical values of these units are identical to energy per area reported as ergs/cm². Older literature describing monolayer studies ordinarily

uses these units. In the mks based SI system of units force per length is reported as Newtons/meter. Numerical values of surface tension and film pressure reported as mN/m (milliNewtons/meter) are exactly identical to values given as dynes/cm.

Marine Microlayer – The term marine microlayer refers to the very thin layer of water near the surface that includes the material found at the air/water interface. This is a habitat for surface-dwelling marine organisms and a region of greatly increased concentration of many organic and inorganic chemical substances. In practice, the method of sampling determines the region described by the term. In much of the recent work describing the marine microlayer, the thin layer of water that will adhere to a glass plate when passed through the interface is being described [79]. The rotating glass cylinder sampling devices used by Carlson, et. al., [580] at Oregon State University makes use of this technique to collect a thin layer of surface water continuously. Under ideal conditions, these devices collect a water layer on the order of 60 μm thick and are the best systems devised to date for continuous field sampling of such a thin layer near the surface. Another commonly used technique for microlayer sampling is to touch a window screen to the surface and then drain the water that is captured between the wires into a collection bottle [79,163,165,179,233,234]. The screen sampler obtains water samples on the order of 500 μm thick. Comparing these thicknesses of water to the less than 10 nm thick film believed to be responsible for the physical effect of capillary wave damping, it can be seen that the screens and glass surfaces collect great quantities of water and other material near the surface relative to the small quantity of material adsorbed at the air/water interface that is actually responsible for damping capillary waves. Therefore, the microlayer may be found to be enriched with materials that have nothing to do with the presence of a natural surface-active film or of the existence of a visible slick.

UNDERLYING PHYSICAL PRINCIPLES AND SELECTED SAMPLE DATA

Many of the behavioral characteristics of pure surface-active compounds studied in the laboratory are related to the behavior of sea slicks, because studies of slicks by chemical oceanographers and environmental chemists have shown that the damping of capillary waves is more accurately attributed to naturally-occurring surface-active chemical films than to spilled petroleum. A large body of literature exists in the field of surface chemistry that has been generated since the turn of the century and that relates specifically to the properties of insoluble monomolecular films and adsorbed monomolecular films at the air/water interface. A cursory examination of the surface chemistry literature immediately provides some interesting pertinent information.

Equations of State

Single-molecule-thick films can be formed by a surface-active compound, because one end of the molecule forms a hydrogen bond with the substrate water, while the remainder of the molecule is hydrophobic and usually less dense than water. Such a material is autophobic—it will not spread spontaneously over its own monolayer. Since film pressure is defined as the difference between the surface tension of water and the surface tension of water covered by the film, a material of this type can be modeled as a 2-D gas on the surface of water. The simplest equation of state for a monomolecular film is a direct analog of the familiar ideal gas law for 3-D gases [210,404].

Film Pressure: $\pi = \gamma_w - \gamma_f$ (1)

Ideal gas: $PV = NRT$ (2)

Ideal monolayer: $\pi A = nkT$ (3)

Natural films: $(\pi + b) A = xnkT$ (4)

or: $\pi A = c_0 + c_1\pi + c_2\pi^2$ (5)

where

π = film pressure	R = gas constant
γ_w = surface tension of water	T = temperature (°K)
γ_f = surface tension of film-covered water	A = area covered
P = pressure	k = Boltzmann's constant
V = volume	n = no. of molecules, and
N = no. of moles	$b; x; c_0; c_1; c_2$ = constants.

Several empirical equations of state can be used to describe natural films [36]. Equation (4) is useful for more accurate representation of the low-pressure region. Equation (5) gives a general representation of the entire pressure-area isotherm, and it is more useful for the high-pressure regions. Data for many natural films collected from the waters of the North Atlantic and from the Chesapeake Bay have been summarized by reporting the constants for Eq. (5) [35,36].

The equation of state information defining the properties of a surface film is most often presented as a graph of the film pressure vs the area covered by the film. As the film is compressed to smaller areas, the pressure rises such as in the compression of a gas. Data of this type for over a hundred natural films that were collected from many locations at sea have been gathered during the past five years and will be presented elsewhere. A single natural sample from this group, which has been characterized physically in detail, will be discussed in this report as an example.

Laboratory Data for a Natural Film Compared to Pure Monolayers

A detailed research paper on laboratory measurements of surface films is beyond the scope of this report. Much more information is contained in the attached references. Data measured in this laboratory for three samples are presented to provide examples for discussion. Monomolecular films of the pure compounds stearic acid and oleyl alcohol are compared to the natural film that formed on the surface of a sample of seawater. The seawater sample was collected from 0740 to 0800 on February 1, 1989, at 33°58'N, 119°37'W during a field experiment conducted south of Santa Cruz Island off the California Coast. Figures 1 through 4 present the results.

At the time of collection, the seawater sample was poisoned with sodium azide to stop biological activity and was stored in the dark until analyzed. It is better, of course, to analyze such samples immediately because of the possibility that the material may change before analysis. However, because of the kind of instrumentation needed to make the measurements being discussed, this is not always possible. Langmuir film balance equipment cannot be used aboard ship, but many seawater samples have been measured within minutes or hours of collection by ferrying the water to shore-based laboratories immediately. Such samples have been compared to many others that have been poisoned and stored for months. Qualitatively the results are remarkably similar. The quantity of surface-active material recovered from stored water samples is believed to be always less than that originally present in the water because of losses to the walls of the containers by adsorption. In any case, we are mostly concerned with the qualitative behavior, since the quantity of material varies from sample to sample. There is nothing unusual about the sample selected for use in this report as an example.

The 3-l seawater sample was stirred and divided into an 800 ml and a 2200 ml fraction. The film that formed on the 800 ml component was compressed in a Langmuir trough to determine the pressure vs area isotherm, the surface potential change vs area isotherm, and the surface compressional modulus as a function of area. The compression rate was 0.615 cm²/s. Figure 3 shows pressure vs area and surface compressional modulus vs area results along with the damping coefficient measured as a function of area for an equivalent quantity of material. Figure 4 shows the results for the measurement of surface potential as a function of area for this sample that were measured simultaneously with the pressure vs area data. Note that it is not necessary to know the exact quantity of material to determine any of these data.

Monolayer films from known quantities of stearic acid and oleyl alcohol were spread from a chloroform solution onto distilled water and compressed in the same apparatus. Because the quantities of the pure compounds were precisely known, a calculation of the pressure vs area per molecule was possible. From the film pressure vs *area per molecule* data for the oleyl alcohol film, the quantity of material necessary to produce a film pressure vs *area* isotherm for oleyl alcohol that closely matched the natural sample was computed and found to be 0.26 μg. This is one method of roughly estimating the quantity of material in the surface film recovered from the seawater sample (keeping in mind that the original sample may have contained more and that some material undoubtedly remains dissolved in the bulk water). Figure 2 shows the film pressure vs area curve for 0.26 μg of oleyl alcohol. Oleyl alcohol forms a 2-D liquid film. Figure 1 shows for comparison the film pressure vs area data for the same quantity, 0.26 μg, of the 2-D solid film, stearic acid.

Effect of Surface Films on Waves

The Kelvin Equation Relationship of Surface Tension to Wave Velocity

The classical relationship between surface tension and gravity as restoring forces and the velocity of waves on a deep basin is given by the Kelvin Equation [20,191,509]:

$$v^2 = \frac{g\lambda}{2\pi} + \frac{2\pi\gamma}{\rho\lambda} \quad (6)$$

where

v = wave velocity
g = gravitational constant
λ = wavelength

π = 3.14
γ = surface tension, and
ρ = density of water.

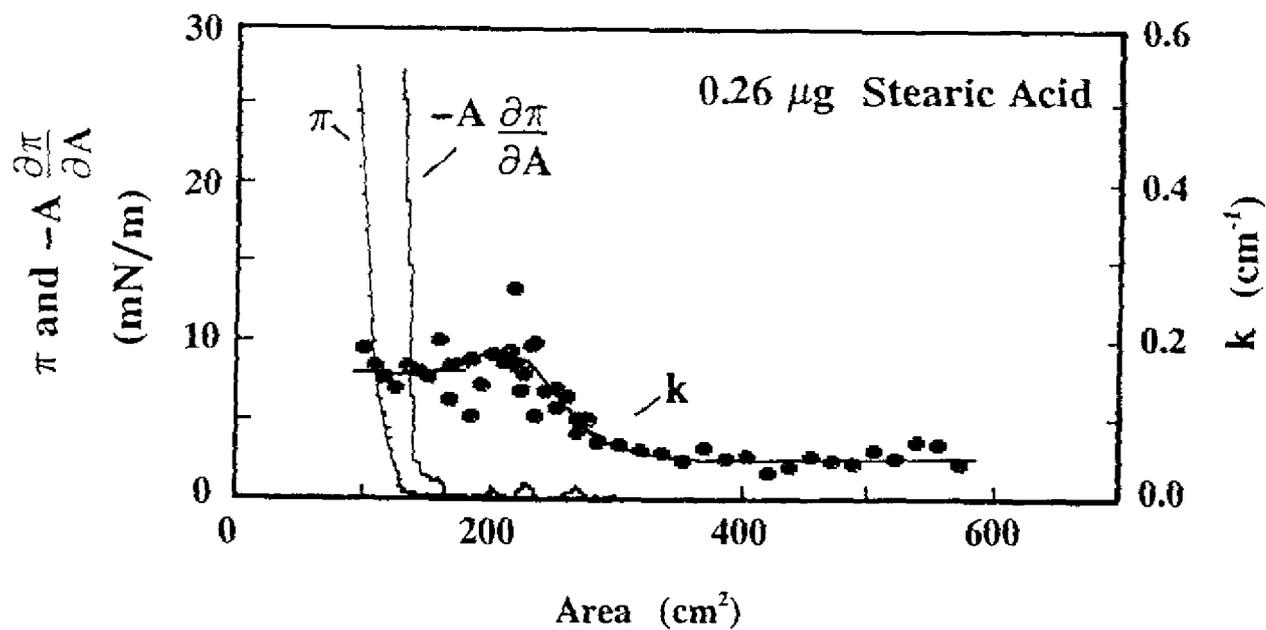


Fig. 1 - Stearic Acid Monolayer: Film pressure π , modulus $-A(\partial\pi/\partial A)$, and damping coefficient k for 30 Hz capillary waves as a function of the area occupied by 0.26 μg of stearic acid

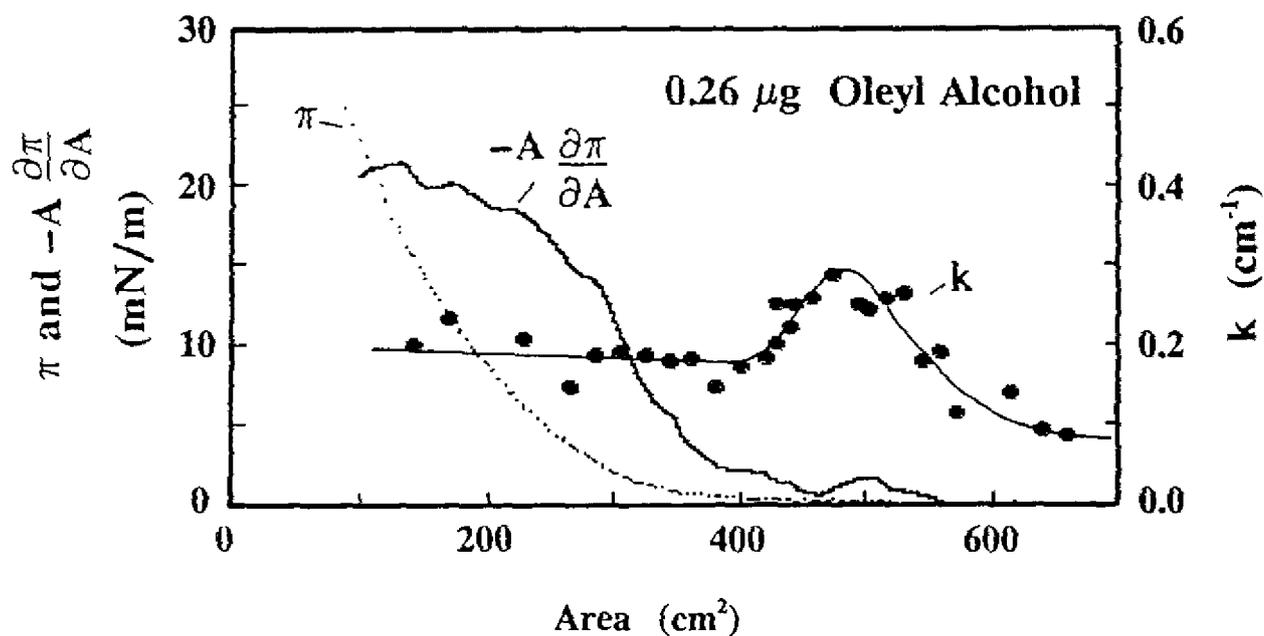


Fig. 2 - Oleyl Alcohol Monolayer: Film pressure π , modulus $-A(\partial\pi/\partial A)$, and damping coefficient k for 30 Hz capillary waves as a function of the area occupied by 0.26 μg of oleyl alcohol

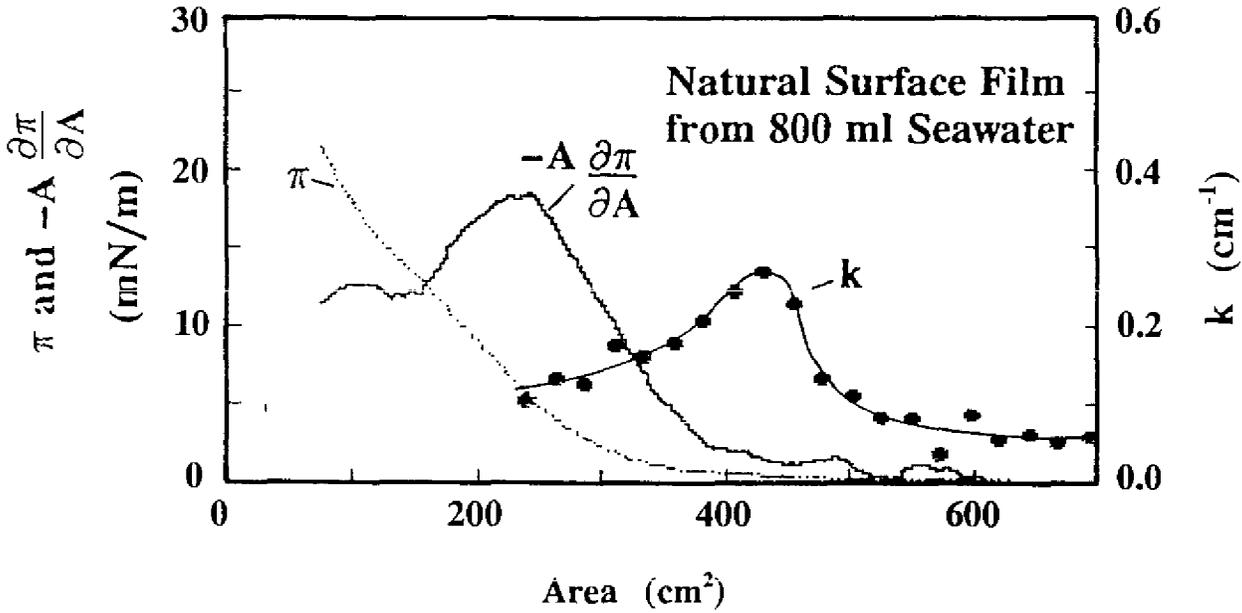


Fig. 3 - Natural Surface Film: Film pressure π , modulus $-A(\partial\pi/\partial A)$, and damping coefficient k for 30 Hz capillary waves as a function of the area occupied by the film formed on the surface of an 800 ml sample of seawater after standing overnight

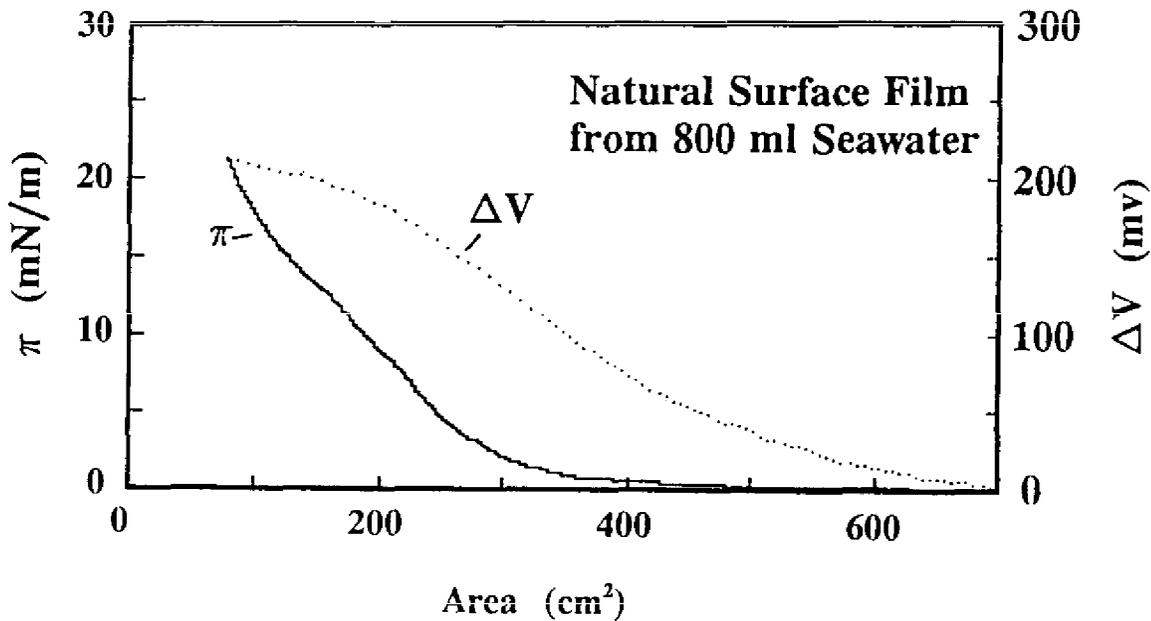


Fig. 4 - Natural Surface Film: Film pressure π and simultaneously measured surface potential change ΔV as a function of the area occupied by the film formed on the surface of an 800 ml sample of seawater after standing overnight

The two terms on the right-hand side of Eq. (6), often referred to as the gravity term and the capillary term, are equal for a wavelength of approximately 1.7 cm. Therefore, capillary waves are generally regarded as waves shorter than 1.7 cm in wavelength. The equation has been shown to give correct results for several monolayer films studied in the laboratory [20].

Capillary Wave Damping

Damping of capillary waves—the effect of the surface-active material that makes slicks visible—can be described by a simple exponential decay constant.

$$A = A_0 e^{-kx} \cos (2\pi x/\lambda) \quad (7)$$

where

A = measured amplitude	π = 3.14
A_0 = initial amplitude	x = distance to measured wave, and
k = damping coefficient	λ = wavelength.

Damping coefficients k for the three samples discussed above were determined by using this equation.

The remaining 2,200 ml fraction of the seawater sample mentioned above was used to fill a glass-bottomed tray in which capillary waves were generated for the determination of the damping coefficient associated with the film. Making the crude assumption that in both cases the seawater was allowed to stand long enough so that all of the surface-active material had accumulated on the surface, the area results for the wave-damping apparatus were normalized to those of the Langmuir trough by the ratios of the volumes of water used in each instrument. In other words, the area values for the wave damping experiment with the natural material were multiplied by the factor $800 \div 2200$ for presentation on the same area scale in Fig. 3. In the cases of stearic acid and oleyl alcohol, since areas per molecule are known precisely as in the case of the pressure vs area isotherms, the normalization is exact and this method is unnecessary. Figure 1 shows the stearic acid damping results, and Fig. 2 shows the results for oleyl alcohol.

Data presented in the figures were obtained by generating 30 Hz capillary waves on the surface of a water sample contained in a 1 cm deep glass-bottomed tray. A video camera connected to an Amiga microcomputer produced digitized images of the damped wave profile that appeared when a horizontal line on the wall behind the tray and below the water level was viewed at a low angle through the rippled surface. Equation (7) was fitted to the digitized wave profile by the method of least squares to obtain values for the damping coefficient k . Motion of the wave image was stopped by a chopper placed in front of the camera. This method produces a recorded image of the entire propagation distance of the wave from its generation point until it is completely damped. Peak and trough locations for 12 to 15 waves along the train were used to determine each of the plotted k values shown in Figs. 1 through 3.

Laboratory studies conducted in the 1960s [162] and the recently measured data presented in Figs. 1 through 4 as examples indicate that the *maximum wave damping effect (maximum k) occurs at very low film pressures*. This means that very small degrees of compression of the surface films that produce small changes in surface tension (< 3 dynes/cm) can result in visible slicks. It is also apparent from the sample data that the magnitude of the damping coefficient is not a simple linear function of the surface

compressional modulus E_s of the film. The surface potential data shown in Fig. 4 prove that a measurable coherent film was present at areas larger than that at which the damping maximum occurred during the compression.

The data presented in Figs. 1 through 3 indicate that a maximum occurs in the damping effect as the concentration of material on the surface increases. This effect has also been observed by others. Shuler and Zisman [586] studied films of polyoxyethylene (Union Carbide POLYOX PEO WSR N10, average molecular weight 90,000) that was spread as a monolayer from a solvent and measured the damping coefficient for 60 Hz waves. Maxima in the damping coefficient were observed on compression of the film to smaller areas. The same compound was studied (at 30 Hz) by Scott and Stephens [460] who found good agreement with the measured data of Shuler and Zisman [586]. They were able to account for the two-peak damping behavior simply on the basis of the material's surface activity without invoking specific intramolecular mechanisms by using the theoretical treatment of Lucassen and Hansen [329,331,527]. The damping of 200 Hz capillary waves on aqueous solutions of fatty acids containing from 5 to 10 carbon atoms was studied by Lucassen and Hansen [331]. All the measured damping coefficients for these compounds passed through a single maximum, as the concentration of material in solution was increased. A graph of these results is also shown on p. 204 of the book by Miller and Neogi [585], which presents a detailed mathematical analysis of capillary wave damping in Chapter V indicating that a maximum in the damping factor is to be expected (See ref. [585], p. 235, problem V-1).

Surface Compressional or Dilational Modulus of Elasticity

A monolayer's resistance to compression can be indicated by the change in film pressure $\partial\pi$, which results from a fractional change in the area confining the film $\partial A/A$. The surface compressional modulus E_s is defined as the ratio of these two quantities:

$$E_s = - \frac{\partial\pi}{(\partial A/A)} \quad \text{or} \quad - A \frac{\partial\pi}{\partial A} \quad (8)$$

which is equivalent to
$$E_s = - \frac{\partial\pi}{\partial \ln A} \quad \text{or} \quad \frac{\partial\gamma}{\partial \ln A} \quad (9)$$

For stiff films E_s is large, while for more elastic films E_s is small and for a clean ($\pi = 0$) surface E_s is 0. For a film described by the equation of an ideal 2-D gas (Eq. 3) E_s is equal to π for all values of area. Classically, for real films studied in a Langmuir trough, the value of E_s has been computed from the film pressure vs area isotherm. For the purposes of this review the value E_s computed in this manner is called the "static" modulus, E_{STATIC} .

In 1872, Marangoni noted that wave attenuation effects might be caused by changes in surface tension brought about by the expansions and compressions of surface films [245,323]. Recent investigators have attempted to incorporate this effect into theories describing wave damping and have argued that dilational modulus values measured under dynamic conditions are more suitable for quantitatively describing wave damping effects on monolayers [244]. In general, surfaces covered by monolayers exhibit viscoelastic behavior and there is an associated surface dilational viscosity η . The paper by Hühnerfuss, Lange, and Walter [244] identifies five relaxation processes that could account for the viscoelastic behavior: (1) diffusional interchange between the undulating surface and the bulk solution

in the case of partially soluble films, (2) lack of instantaneous thermodynamic equilibrium for insoluble films, (3) exchange of monomers with micelles, (4) relaxation through adsorption barrier processes, and (5) exchange with multilayer particles.

The result is a phase difference between the surface area variation and the surface tension variation that the authors account for by introducing a complex modulus (See also Gaines [154], pp. 23-27).

$$E = |E| \cos \Theta + i |E| \sin \Theta \quad (10)$$

where

$$\begin{aligned} \Theta &= \text{the phase difference between } \partial\gamma \text{ and } \partial A, \\ |E| \cos \Theta &= \text{the 2-D elasticity coefficient, and} \\ |E| \sin \Theta &= \text{a surface loss coefficient.} \end{aligned}$$

According to Hühnerfuss, Lange, and Walter [244], the surface loss coefficient $|E| \sin \Theta$ can be related to the dynamic surface dilational viscosity by

$$|E| \sin \Theta = \eta \omega \quad (11)$$

where

$$\begin{aligned} \eta &= \text{the surface dilational viscosity, and} \\ \omega &= 2\pi f, \text{ the angular frequency of the motion.} \end{aligned}$$

Illustrative values of $|E|$, the "dynamic" dilational modulus, are shown under the heading E_{DYNAMIC} in Table 1. The phase shift Θ has been measured for 1 Hz waves in a laboratory wave channel, and the time-dependent surface viscosity was measured in a separate experiment by using a canal viscosimeter to compute the dynamic dilational modulus values $|E|$. Values of $|E|$ are found to be consistently lower than the static values computed from pressure vs area isotherms determined by slow film compressions. Table 1 shows data for three pure surfactants at high film pressures. These data indicate significant differences between the static and dynamic values of the surface compressional modulus except in the case of the ionic surfactant hexadecyltrimethylammonium bromide. The authors suggest that the reason for the difference between E_{STATIC} and E_{DYNAMIC} is that the higher compressional velocity on a wave-covered surface causes significant disturbances in the optimum arrangement of the film molecules, which

Table 1 – Examples of Static vs Dynamic Modulus Data at High Film Pressures [a]

Substance	π mN/m	E_{STATIC} mN/m	E_{DYNAMIC} mN/m	Θ^b deg	η Pa s m
Methyl hexadecanoate	38	81	14.7	130.5	0.00178
<i>cis</i> -9-octadecen-1-ol (oleyl alcohol)	28	71	4.7	-41.0	0.000494
hexadecyltrimethylammonium bromide	30	59	43.2	1.0	0.000120

^a The data are summarized from ref. [244].

^b The phase difference Θ between $\partial\gamma$ and ∂A was determined for 1 Hz waves.

is achieved only when the film remains still for a period of time. Data of this type for natural films have not been measured.

The reader is reminded of two facts about natural films: (1) that significant capillary wave damping takes place at very low film pressures, and (2) natural films are believed to be composed of complex mixtures of materials that are likely to be less regularly organized in a compressed film than the pure single compounds discussed here. Coarse E_{STATIC} values for many natural films given by the reciprocals of coefficients of compressibility reported in Ref. 35 lie in the range 12.4 to 29.9 mN/m. These values that were obtained from least squares best fits of the π vs A data to the empirical equation $\pi = M \ln(A) + B$, where M and B are the mathematically determined constants, are nominal values representing each film over its entire compression range. It is more precise to compute E_{STATIC} for each value of the area and plot it along with the film pressure. E_{STATIC} data determined in this manner for stearic acid, oleyl alcohol, and a natural surface film compressed at a constant rate of 0.615 cm²/s are shown in Figs. 1 through 3. For the 2-D solid film, stearic acid (Fig. 1), the modulus reaches a maximum value greater than 28 mN/m. The sample of oleyl alcohol in Fig. 2 achieved a maximum modulus value of only about 22 mN/m and shows a distinct slope change near the area 290 cm² which corresponds to a film pressure of only about 2 to 4 mN/m. This indicates a slowing of the rate of increase of stiffness with decrease in area and may be caused by a change in configuration of the surface molecules as they begin to be more closely crowded together. In the case of the natural film sample (Fig. 3) there is a sharp break in the modulus curve at about an area of 250 cm², which corresponds to a film pressure of about 4-5 mN/m. Most likely this represents an actual buckling or collapse of the film or squeezing out of lower molecular weight or less strongly adsorbed molecules from the mixture. This "collapse" phenomenon has been observed in many of the natural films studied in our laboratory.

Vertical Transport of Surface-Active Material

Soluble Surfactants and the Gibbs Adsorption Isotherm

The hydrophil-lipophil balance of some surface-active compounds is such that they can be dissolved (in low concentrations) but will preferentially adsorb at the air/water interface. In such cases, a monolayer at the surface is in equilibrium with a low concentration in solution. The Gibbs Adsorption Isotherm describes the relationship between the concentration at the surface and the change in surface tension of such a solution with concentration of the surfactant [404].

$$\Gamma = -\frac{1}{RT} \frac{\partial \gamma}{\partial (\ln c)} \quad (12)$$

where

- Γ = moles/cm²
- RT = gas const. & °K
- c = moles/liter, and
- γ = surface tension.

Pure soluble surfactants are customarily characterized by plotting surface tension vs log concentration to obtain an isotherm that contains a linear segment of slope $-RT\Gamma$. The area per molecule of the adsorbed film can be obtained from Γ (moles/cm²) since there are 6.023×10^{23} molecules/mole and 10^{16} Å²/cm². The relationship between the surface tension and bulk concentration of natural surface-active material in seawater is unknown because the concentration of the surface-active material is unknown. This relationship cannot be easily determined with present technology because of the complex nature and low

concentration of the natural film-forming material that, of course, is present within a bulk solution containing mostly nonsurface-active organic matter. Studies with appropriate model compounds (of known high molecular weights) compared to the empirical behavior of natural films would be useful here.

Energy Associated with Adsorption or Desorption

Once a molecule leaves the bulk water by adsorption at the air/water interface, a considerable amount of energy is required to put it back into solution (810 cal/mole per CH₂ group [404]). As an example, for a compressed stearic acid monolayer occupying an area of 25 square Angstroms per molecule, there are 17 CH₂ groups. Since there are 6.02 x 10²³ molecules/mole, 10¹⁶ A²/cm², and 4.18 x 10⁷ ergs/cal, the above number converts to 382 ergs/cm². This may seem small, but when the extreme thinness of the film being discussed is considered ($\approx 25 \times 10^{-8}$ cm), it can be seen that the energy requirement is substantial, since $382 / (30 \times 10^{-6}) = 1.53 \times 10^9$ dynes/cm²; since 1 atm is 1.01 x 10⁶ dynes/cm², this is equivalent to $\approx 1.5 \times 10^3$ atmospheres. It is believed that numbers for natural films would not be quite so high, but qualitatively the result is similar.

Rates of Adsorption at Interfaces

For adsorbing surface-active compounds, the rate of change of the number of molecules at the surface with time can be described by the following general equation [404]:

$$\frac{\partial n}{\partial t} = Bc(1-\Theta) \quad (13)$$

where

- n = no. molecules
- t = time
- B = constant
- c = bulk concentration, and
- Θ = fraction of surface covered.

In a system of constant area this can be combined with $\partial n/\partial t$ obtained from the combination of Eq. (1) with either Eq. (3) or Eq. (4) to give a relationship involving the change of surface tension (γ) with time:

$$\frac{\partial \gamma}{\partial t} = - \frac{BkTc(1-\Theta)}{A} \quad (14)$$

The consequence of these equations is that when the surface is clean, adsorption is fastest and proportional to the bulk concentration near the surface. As material loads the area of the surface, the adsorption rate slows considerably. Data of this type are very sparse for natural films, however some pure compounds have been carefully studied [404]. For octyl alcohol solutions 3.44 x 10⁻³ mol/l, a surface tension change of approximately 27 dynes/cm was measured in 10² s by the oscillating jet method. Radioactive tracer studies of sodium stearate solutions 3.26 x 10⁻⁶ mol/l showed measurable increases in adsorption continuing over a period of 3 h.

Transport by Bubbles

In addition to the mechanism of direct adsorption from the bulk water to the surface, adsorption at the air/water interface of rising bubbles can transport significant amounts of surface active material rapidly to the surface (see cross index references under key word **bubbles**). This mechanism greatly increases the rate of transport of surface-active material to the sea surface and may contribute significantly to the production of slicks observed behind moving ships. Upon the bursting of the bubbles, the material from the bubble's (internal) air/water interface is merged with the material already directly adsorbed on the sea surface. Under low wind conditions lacking whitecapping, no corresponding downward transport mechanism exists. Under rough conditions with much whitecapping, not only can material be carried downward, but stripping of surface-active material from the sea surface and into the marine atmosphere where it is associated with the marine aerosol can also be a significant removal mechanism. Marine particulate matter can also be formed because of the accumulation of high molecular weight compounds at bubble interfaces.

Horizontal Transport of Surface-Active Material*Thermodynamics of Spreading Monolayers and Energy of Spreading*

Energy associated with a compressed monolayer will cause it to spread spontaneously over a clean water surface. The thermodynamics of spreading of monolayers at the air/water interface has been well studied for some pure compounds. The treatment of Harkins [210] is summarized here; for an alternative treatment see Osipow [404]. Energies are associated with two conditions: "spreading" and "extension."

Spreading – When the moving boundary of a monolayer spreading over water increases the area of the film by 1 cm² and simultaneously decreases the area of clean water by 1 cm², the energy of the film is increased while the energy of the water is decreased. The net result is the energy of spreading, H_S. This is the case most often encountered.

Extension – It is possible to increase the area of the film without spreading it over a new clean water surface, for example, by lifting a wetted ring at the interface slightly above the surface. The associated energy is called the energy of extension, H_E.

Harkins [210] has considered the case for pentadecylic acid (pentadecanoic acid) at 20° C and a film pressure of 3 dynes/cm. The surface tension of clean water is taken as 72.75 dynes/cm, therefore the surface tension of the film-covered water is 72.75 – 3.0 = 69.75 dynes/cm, which is equivalent to 69.75 erg/cm². The change of surface tension with temperature for a monolayer of this material is known to be –1.106 dynes/(cm °K). The change in surface tension of clean water with temperature is –0.148 dynes/(cm °K).

The extended film's free energy is 69.75 erg/cm². The extending film also absorbs heat Q_E, which must be accounted for when determining the total energy.

$$\text{Extension:} \quad H_E = \gamma_F - T(\partial\gamma_F/\partial T) \quad (15)$$

$$\text{Spreading:} \quad H_S = H_{E(\text{FILM} + \text{WATER})} - H_{E(\text{WATER})} \quad (16)$$

Pentadecanoic acid monolayer at 20 °C and $\pi = 3.0$ dynes/cm:

$$\begin{array}{ll} \text{(a) for film covered water:} & H_E = 69.75 - 293 (-1.106) = 394 \text{ ergs/cm}^2 \\ \text{(b) for clean water:} & H_E = 72.75 - 293 (-0.148) = \underline{116} \text{ ergs/cm}^2 \\ \text{Total energy for spreading,} & H_S = 278 \text{ ergs/cm}^2. \end{array}$$

To make this calculation it is necessary to know the change in surface tension of the film in question as a function of temperature. A complication for some films is that different phases exist in the monolayer, and the slope of surface tension vs temperature changes abruptly when different phases exist. Thermal studies of natural sea surface films have not been done.

Shear Stress on Monolayers and Kinetics of Spreading Caused by Surface Tension Gradients

Compression of Surface Films by Wind

A shear stress can be exerted on a water surface by a flowing gas stream or by the wind. According to Davies and Rideal [108, p. 267] and to Roll [434, p. 152], the tangential shear stress is generally considered as constant with height in the first few meters of the boundary layer, and for wind velocities below about 5 m/s the following relationship holds:

$$\tau = \rho C_z v_z^2 \quad (17)$$

where

$$\begin{array}{l} \tau = \text{stress (dynes/cm}^2\text{)}, \\ \rho = \text{density of air (gm/cm}^3\text{)}, \\ C_z = \text{drag coefficient (dimensionless), and} \\ v_z = \text{velocity at height } z. \end{array}$$

C_z is a proportionality constant dependent upon the height at which the wind velocity is measured. Davies and Rideal report values for C_z of 0.0009, when z is several meters and 0.0025 when z is 5 cm. These figures apply to aerodynamically smooth surfaces irrespective of whether or not a film is present. In the fields of physical oceanography and marine meteorology, it is customary to report the values based on wind measurements corrected to 10 m above the surface. The book by Roll [434] contains a large table of C_{10} values determined by various methods that range from 0.0004 to 0.0062. A value in the range 0.0011 to 0.0015 was determined by the more recent and presumably more accurate wind profile measurements when only neutral conditions were considered [434]. The importance of the drag coefficient in air-sea interactions is also discussed in a recent paper by Geernaert [582].

The stress at the air/water interface in the x direction can be opposed by a monolayer being compressed by the wind. The natural tendency of a compressed monolayer is to spread spontaneously toward cleaner water until no further gradient in the surface tension exists. In a rectangular channel of width w , wind blowing over a monolayer film compresses the film downstream to smaller and smaller areas wx until the surface pressure gradient $\partial\pi/\partial x$ is equal to the shear stress τ . This leads to the force balance equation:

$$\frac{\partial \pi}{\partial x} = \rho C_z v^2 \quad (18)$$

This relationship has been checked experimentally. Six pure surface-active compounds of known equilibrium spreading pressures were studied in 1970 to determine the wind velocity necessary to compress their monolayers to a length of 20 cm in a bench-scale small wind tunnel [169]. A 10 x 40 cm rectangular tray approximately 0.3 cm deep was placed in a 11.3 cm diameter acrylic tube through which air was drawn by a variable speed fan. A loose, floating, paraffin-coated thread was attached across the surface of the water in the trough 20 cm from one end. An excess of material required to form a monolayer in one half of the tray (20 mg) was added to the downwind side of the thread. With no wind the thread bowed toward the direction from which the air would flow. The wind velocity was gradually increased until the thread became slack at the point where the drag caused by the wind just balanced the spreading force caused by surface tension gradient. Table 2 shows the results along with a determination of C_z from Eq. (18).

Table 2 - Drag Coefficients Computed from Wind Velocities Balancing Equilibrium Spreading Pressures of Several Pure Monolayers

Substance	π_{ESP}^a (mN/m)	Wind Velocity (m/sec)	Drag Coefficient ^b (dimensionless)
Tricresyl Phosphate	9.0	5.62	0.0012
Cottonseed Oil	13.8	7.51	0.0010
Glyceryl Trioleate	17.0	8.02	0.0011
Oleyl Alcohol	32.9	8.60	0.0019
Sorbitan Monooleate	40.8	8.71	0.0023
Dodecanol-1	41.1	9.26	0.0020
		Mean:	0.0016

^a π_{ESP} = Equilibrium Spreading Pressure = Film pressure of the monolayer in equilibrium with bulk material.

^b The drag coefficients C_z were computed from Eq. (18) by using a monolayer length, x , of 20 cm and an air density of 0.001185 gm/cm³.

Spontaneous Spreading of Monolayers in the Absence of Wind

Equation (18) can also be used to describe the spontaneous spreading of monolayer films toward regions of cleaner water that results from a surface pressure gradient. In such a case, since the monolayer is coupled to the air/water interface, water is dragged below the moving monolayer, and therefore the appropriate density to be used in the equation is that of water. Schulman and Teorell reported in 1938 that an underlying layer of water \approx 0.01 mm thick can be dragged along below a

moving monolayer of oleic acid [108,457]. The assumption is that the monomolecular layer of polar organic material at the surface is very much thinner than the associated water layer, and therefore the influence of the density of the compound is negligible. Since the parameter of interest in this case is the velocity of the leading edge of the monolayer, Eq. (18) can be rearranged to give

$$v_0^2 = K \frac{\partial \pi}{\partial x} \quad (19)$$

where

$$\begin{aligned} v_0 &= \text{water velocity at the interface (cm/sec),} \\ K &= 1/\rho C_z, \text{ a constant (cm}^3/\text{g), and} \\ \rho &= \text{density of water, 1 g/cm}^3. \end{aligned}$$

The velocities of spreading of several pure surfactants on the surface of distilled water were measured in our laboratory in a thermostatted channel 3.5 m long by 10 cm wide. All of the compounds investigated were liquids at room temperature. A single drop (20 mg) of the bulk compound was placed on the water surface inside a 5-mm-high section of a 3-cm (i.d.) metal tube that was suspended in the interface to provide a confined circular area. The autophobic surfactants did not spread over their own monolayers, so the several millimeter diameter bulk liquid drop was surrounded by its monolayer at the equilibrium spreading pressure within the confining ring. A Wilhelmy Plate surface tension sensor was suspended in the air/water interface at various distances from the confining ring. The ring was dropped through the interface, and the time was recorded for the first detection of surface tension change caused by the leading edge of the spreading monolayer film. Table 3 shows the results of these runs plus computations of the constant K corrected for temperature and the corresponding values of C_z . The values of K (and of C_z) seem to be influenced more by the structure of the hydrophilic head groups than by the hydrocarbon tail lengths. The head groups would be expected to dominate the interaction with the underlying water.

Surface Current Velocities Determined from Surface Tension Gradients

Equation (19) can be used to calculate the velocity of water at the air/sea interface when the gradient of surface tension and the constant K are both known. In 1937, N. K. Adam [1] described a method of determining the surface tension of the sea *in situ* by dropping a series of calibrated oils (See the listings in the cross index under the topic *spreading oils*). Recently a towed instrument was used to make very high-resolution measurements of surface tension by automatically dropping a series of Adam's spreading oils as the device was towed across the wakes of several ships. Results of this experiment were described by Peltzer, et al., [414]. Repetition of the data from the numerous crossings will not be made here, but, in general, two strong bands of reduced surface tension were associated with the edges of the wakes. Additional regions of reduced surface tension were also commonly seen within the wake, however no systematic pattern was immediately obvious. Coming from the undisturbed water into each of the edge bands, a gradient of surface tension decreasing from the clean water value to a low value was observed followed by a gradient rising back to the clean water value. Based on Eq. (19), a sustained pattern of converging surface currents caused by the passage of the vessel would explain the surface tension gradients. This suggests several counter-rotating water circulation patterns that persist in the wake. The magnitudes of the surface currents acting at right angles to the wake could be computed from Eq. (19) if the value of K for natural films were known. The best that can be done at present is to

Table 3 - Surface Film Spreading Speed Data For Several Pure Surfactants

Surfactant	π_{ESP} (dynes/cm)	x (cm)	t (s)	Temp. (°C)	K^* (cm ³ /gm)	K_{25} (cm ³ /gm)	C_z
POE(4)Lauryl Ether	44.5	300	22.1	25.0	1242	1242	0.00081
PEG(200)Monolaurate The equation for the straight line describing the temperature effects for the 300 cm runs at the right is $K^* = 20.705 \times \text{°C} + 676.45.$	43.0	350	30.5	20.0	1072	1175	0.00084
		300	23.5	25.0	1137	1137	
		300	23.9	21.9	1099	1163	
		300	23.5	20.0	1137	1240	
		300	24.0	17.1	1090	1253	
		300	23.0	13.7	1187	1420	
		300	25.8	13.2	937	1180	
		300	26.3	13.1	908	1153	
		300	27.2	9.8	849	1162	
		300	26.6	9.8	887	1201	
		250	17.8	20.0	1147	1250	
		200	12.9	20.1	1118	1219	
		150	8.5	20.0	1086	1189	
		100	4.8	20.2	1009	1108	
			Mean:		1192		
PEG(200)Dilaurate The equation for the straight line describing the temperature effects for these runs is $K^* = 20.542 \times \text{°C} + 564.40.$	39.8	300	25.3	25.0	1060	1060	0.00093
		300	28.3	15.2	847	1049	
		300	27.3	13.0	910	1157	
		300	27.1	12.9	924	1173	
		300	30.8	11.2	715	999	
		300	29.3	11.0	790	1079	
		300	30.7	9.8	720	1033	
					Mean:		
PEG(400)Dilaurate	42.2	300	24.0	25.1	1116	1114	0.00091
		300	29.1	8.7	759	1095	
					Mean:		
Sorbitan Monolaurate	45.2	300	27.6	25.0	784	784	0.00128
PEG(400)Diolate	40.8	300	24.3	25.0	1121	1121	0.00089
PEG(400)Monooleate	40.3	300	27.3	24.6	899	907	0.00110
Sorbitan Triolate	40.0	300	27.8	24.8	873	878	0.00114
Sorbitan Monooleate	42.2	300	27.6	25.0	840	840	0.00119
Oleyl Alcohol <i>cis</i> -9-octadecanol	33.5	350	63.7	20.0	315	418	0.00236
		300	43.5	25.0	425	426	
		150	14.5	19.9	479	584	
		100	13.8	19.8	157	264	
					Mean:		

Notes:

- π_{ESP} = Equilibrium Spreading Pressure; values from Garrett and Barger [174].
- K^* values are solutions to Eq. (19) at the stated temperature using x/t values from the table for v_0 and π_{ESP}/x values for $\partial\pi/\partial x$.
- K_{25} values were obtained by correcting from the reported K^* values to 25°C by using 20.6 units/°C.
- Laurate = dodecanoate; Oleate = *cis*-9-octadecenoate; PEG(200) = polyethylene glycol (average molecular weight 200); POE(4) = polyoxyethylene (4 oxyethylene units; each of molecular weight 44).

assume that K lies between about 500 and 1200 based on the values for pure surfactants presented in Table 3. For example, if K is 1100, the measured gradient of 0.02 dynes/cm² along one edge of a wake 3.74 km behind a ship moving at 25 knots (12.9 m/s) leads to a convergence velocity in one region of the wake of approximately 5 cm/s. The validity of this suggested new method of determination of surface current should be confirmed by some alternate method of surface current measurement carried out simultaneously in the field.

Persistence of Films Once Formed

As surface-active material accumulates in one spot on the surface, the surface tension can be reduced considerably at that spot. This fact has been confirmed by numerous *in situ* measurements with spreading oils. A surface-tension-driven, spontaneous horizontal spreading toward areas of cleaner water results from the accumulation of excess material in one area, as indicated by the sections discussing thermodynamics and kinetics of spreading of monolayers. In systems (like the sea) where low concentrations of material are believed to always contaminate the surface, the spreading film herds the widely separated ambient molecules into a surrounding film that opposes and slows the spreading of the film from the point source. Cohesive forces within the surface film oppose total disintegration of the film after it has spread to the point where a surface tension gradient no longer exists. Most likely this is because the surface films are composed of polar organic compounds. Figure 4 indicates that an approximately 200 mv potential difference resulted from compression of the natural sample. This is interpreted to be caused by the change in the orientation of the dipoles associated with the molecules of the surface film being compressed. Figure 5 provides an extremely oversimplified model to illustrate how the dipolar nature of the film material can contribute to its cohesion. When the film is compressed (Case I) the oriented dipoles contribute to the respreading of the film because they repel each other electrostatically. The force associated with this repulsion is included in the surface tension value because of the method by which surface tension is measured. When the film is expanded, the net dipoles are effectively reoriented as if they were horizontal at the surface. Surface potential difference from clean water measured in the vertical direction is zero. However, some equilibrium condition exists between Case III and Case I where the electrostatic attractive forces between the partial negative charge on head of one molecule and the partial positive charge on the tail of another are balanced. The expanded film presumably exists in this condition of surface density until destroyed by stronger mechanical forces. From the surface potential data it can be seen that this condition occurs at areas where the film is so expanded that the surface tension is essentially identical to that of clean water. Also, the reader should not forget the considerable amount of energy required to force a surface-active compound back into solution. This has been discussed under the topic "Energy Associated with Adsorption and Desorption."

Model Compounds to Simulate Natural Films in Laboratory Studies

Early studies of natural surface films often compared them directly to simple fatty acids or fatty alcohols. Such studies of simple systems are very useful to gain insight into likely behavior of the natural material. However, as a result of studies carried out in the 1980s [36], it is known that the early model compounds used for comparison were not the best simulants of natural film behavior. Most laboratory studies of monomolecular films have involved the use of simple molecules of molecular weights on the order of 300. Today, as a result of recent studies of natural films, it is believed that the natural film-forming material consists of an extremely complex mixture containing much higher molecular weight compounds, with a broad distribution of molecular weights and an average on the order of 3000. Polymeric compounds containing many oxygens and multiple long-chain hydrocarbon tails seem to be indicated as better models for the natural material. This size difference from earlier models alters our

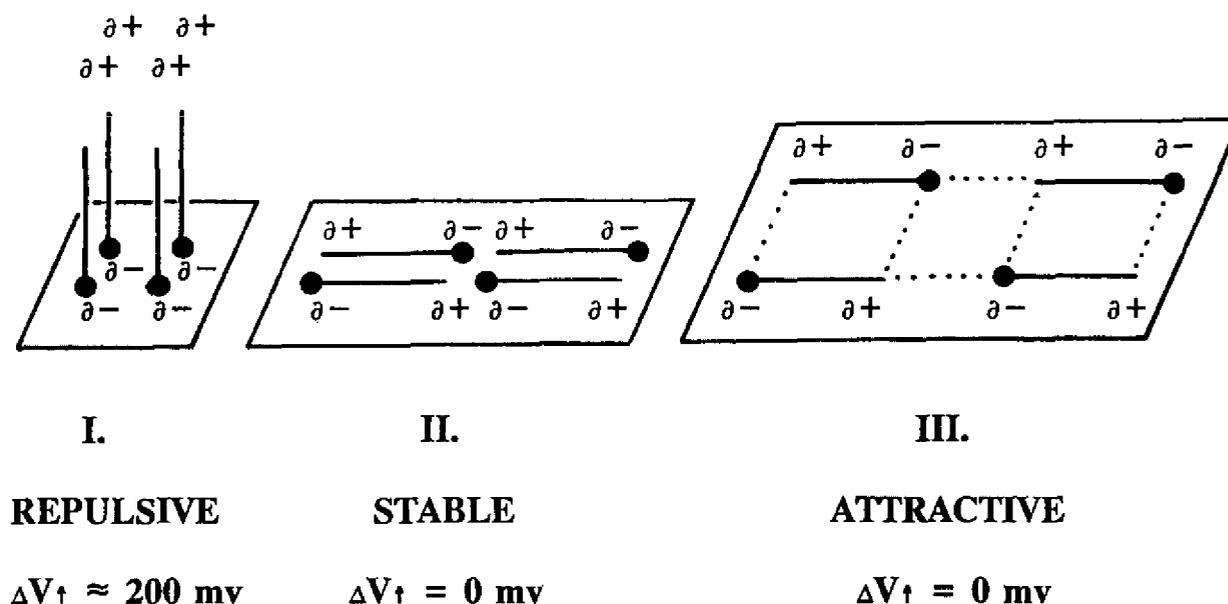


Fig. 5 - Stability of films can be influenced by electrostatic effects caused by the different alignments of dipoles. In a highly compressed film (Case I) the dipoles are forced into a repulsive configuration. When the film is greatly expanded (Case III) partial charges on the dipoles lead to attractions between the film molecules. There is some intermediate condition (Case II) in which the film is stable.

expectations of the influence of the compounds on properties such as diffusion rates and surface viscosities. Information on appropriate model compounds is woefully inadequate to allow unquestioned interpretations of the observations made during field experiments. However, experiments with simple compounds should continue to be conducted in the future, along with materials of higher molecular weight, to tie the results to the previous literature.

CONCLUSIONS

The brief summaries of information contained in this report have been presented as an example of the type of information on topics related to the formation and disappearance of observed regions of damped capillary waves on the sea surface to be found already in the literature and are intended only as a brief overview and guide to the more detailed information to be found in the original papers. The main value of gathering an overview of pertinent information is to identify specifically those measurements on slick-forming materials that are needed to remove gaps in the data that hamper our thorough understanding of sea slick behavior. Several obvious gaps in the data concerning natural slicks on the ocean have been pointed out, and in cases where data do exist, the quantity is small. Measurements of the modulus of elasticity for natural films need to be made under dynamic conditions. The surface dilational viscosity has not been measured. The constant B in Eqs. (13) and (14) that describes the rate of adsorption at the air/water interface is unknown. The change of surface tension with temperature that would permit computation of the total energy for spreading by using Eqs. (15) and (16) has not yet been measured for natural films. An estimate of the drag coefficient C_z relating surface water convergence

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velocities to surface tension gradients can be made from data presented in this report, but a value determined by experiments with natural films would be better. Hopefully this review will inspire future laboratory and field studies pertinent to the effect of surfactants on the physics and chemistry of the air/sea interface. The qualitative behavior of surface films on the ocean has been recognized for many years. What is missing is a sufficiently large set of numerical values specific to natural films or to model compounds that accurately represent the complex mixture actually found at the sea surface.

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CROSS INDEX TO REFERENCES BASED ON KEY WORDS

Numbers under key words indicate references in the following list that contain information about that topic.

ADSORPTION at interfaces: 39 67 95 101 115 131 201 211 255 261 265 266 267 304 333
369 398 404 435 437 439 444 456 458 491 501 531 577 581

AIR-SEA INTERACTION related to interfacial phenomena: 20 22 27 33 51 52 53 62 96
97 111 113 152 178 179 181 221 223 236 292 334 425 515 516 566 570 574 576 582

ANALYSIS of the surface microlayer and air/water interface: 3 11 12 14 15 19 21 23
24 30 31 34 35 36 37 38 45 46 49 53 58 59 60 61 78 79 80 81 82
83 89 95 101 104 105 107 122 124 132 135 136 140 151 152 153 157 159 161 163 165
175 179 184 187 188 192 200 201 205 206 213 215 216 217 219 222 231 233 234 253 254
255 256 265 268 273 275 279 280 281 288 290 311 315 318 319 320 340 341 354 355 356
357 358 364 365 370 371 374 377 378 381 382 383 384 386 389 393 394 395 397 398 402
403 405 406 408 411 419 420 424 425 428 429 430 433 440 451 453 470 474 477 478 479
480 482 483 484 485 486 487 495 500 501 504 512 519 520 521 523 530 531 541 545 552
553 554 555 556 557 558 559 571 576

BUBBLES: 5 18 38 39 40 50 51 52 53 54 76 104 105 112 113 114 121 125
140 155 156 164 166 211 221 230 285 286 287 289 310 316 337 338 339 340 357 372 373
375 379 380 391 407 409 412 413 422 426 427 436 443 444 481 489 503 504 505 510 511
515 536 540 542 543 544 572

DAMPING of capillary waves and short wavelength water waves: 8 43 44 47 57 69 70 91
102 110 121 127 128 129 133 146 148 158 160 162 168 190 191 193 194 202 203 204 227
241 242 247 249 250 259 271 282 300 309 314 324 326 327 328 329 330 331 332 334 346
347 348 352 366 376 388 416 455 456 459 460 462 463 475 507 508 509 514 527 528 532
537 546 549 584 585 586 587

DRAG and wind influence on surface water: 66 108 169 434 457 498 582 591

EVAPORATION RETARDATION by surface films: 41 44 55 149 170 171 189 198 220 226
262 264 296 321 343 350 352 388 423 518 538 563

GENERAL SURFACE CHEMISTRY: 59 77 106 108 109 147 183 185 186 187 197 205 212
226 230 254 260 273 279 303 319 320 322 333 334 337 382 383 404 437 496 500 503 544
547

HYDRODYNAMICS: 48 72 392 414 490 583 585 590

INTERNAL WAVES: 13 137 138 298 299 308 313 363 446 472 573 574 575 588 589

MICROLAYER at the air/water interface: 12 19 37 45 46 53 59 78 79 80 81 82
83 89 95 101 104 105 107 122 124 132 133 135 140 150 151 152 153 161 163 165 179
181 187 188 200 205 206 212 213 214 215 216 217 219 222 223 231 233 234 240 253 254
255 256 270 275 279 280 281 288 290 311 318 319 320 322 324 335 340 341 349 354 355
356 364 367 377 378 383 384 393 394 395 402 405 406 408 411 420 425 440 464 465 470
477 479 480 482 504 512 519 520 522 530 531 535 541 545 548 552 553 554 555 556 557
558 559 570 571 576 578 580 587

MICROWAVES: 7 9 56 229 247 476

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MONOLAYERS at the air/water interface: 2 7 25 26 29 33 35 36 41 42 43 54
 55 56 57 67 77 91 104 108 115 118 119 120 127 129 130 154 158 160 168 169 170
 172 173 174 177 181 183 185 186 198 201 207 212 220 223 225 225 235 236 238 239 240
 241 242 247 248 249 250 251 252 260 261 262 263 264 265 266 267 270 296 303 304 305
 306 307 324 329 332 333 335 344 345 349 350 369 388 400 401 404 409 435 438 441 442
 448 454 457 460 464 465 491 496 514 517 518 535 537 538 559 563 570 586 591

OIL (petroleum or hydrocarbon oils): 1 9 25 28 29 56 58 87 88 118 136 139 141
 142 143 169 172 174 176 199 225 229 232 251 258 272 294 295 307 312 321 342 359 376
 386 387 390 423 432 463 492 577

RADAR: 7 9 13 32 56 71 94 182 196 199 224 229 246 247 248 272 363 471 472
 476 526 533 534

REMOTE SENSING of the sea surface: 7 9 32 41 42 56 63 71 88 93 94 96
 97 133 136 153 180 181 182 196 198 199 212 223 224 229 237 238 240 243 246 248 251
 269 270 272 277 290 297 342 349 359 362 396 446 464 465 467 468 469 471 473 476 493
 494 524 525 526 533 534 535 559 570

SLICKS: 7 9 10 13 14 20 22 25 26 27 28 29 31 32 33 34 35 36
 41 42 52 56 63 66 71 82 83 87 88 94 97 99 118 123 127 133 136 137 138
 141 142 143 148 149 150 151 153 157 159 161 162 163 165 167 171 172 173 174 178 180
 181 185 198 199 200 205 212 214 222 223 224 229 231 233 234 236 237 238 240 241 242
 243 246 247 248 249 250 251 252 253 258 262 265 266 267 268 269 270 272 275 281 283
 290 291 295 296 298 299 304 305 311 319 322 324 328 334 335 336 342 345 349 353 356
 359 362 363 366 367 385 386 387 390 396 409 414 416 417 423 431 432 434 451 452 459
 461 462 463 464 465 466 467 468 469 470 471 472 475 477 492 493 494 499 502 512 518
 519 525 530 531 535 545 548 551 556 558 559 567 570 573 574 578 579 580 588 589 590

SPREADING OILS (for measuring surface tension *in situ*): 1 28 179 197 225 294 307 414 448
 450 577

SPREADING RATES of monolayers and slicks: 4 6 16 33 64 65 68 74 75 84 85
 86 100 109 111 119 120 139 141 142 143 146 149 169 172 174 176 177 179 197 207 209
 226 228 232 257 278 296 302 312 350 351 360 361 399 400 401 414 415 421 438 439 441
 442 445 449 454 457 497 513 517 539

SURFACE POTENTIAL: 2 252 531 556

SURFACE TENSION: 1 106 128 129 208 274 301 304 312 336 346 418 435 474 502 546
 547 550

SURFACTANTS: 8 21 23 24 30 31 34 36 54 76 95 104 105 109 133 150 162
 164 166 181 197 220 223 224 240 255 257 265 266 267 268 290 322 328 332 333 334 335
 349 369 414 444 451 458 459 461 464 465 475 489 491 507 508 514 527 549 559 570 581

TEMPERATURE (including microwave; radiometer; IR; internal waves, etc.): 7 9 56 137 138
 180 195 198 262 263 264 298 299 344 353 410 447 476 488 574 575

WAVES: 8 13 17 96 98 109 126 133 193 249 250 259 271 291 304 308 313 334
 368 417 434 461 463 472 490 498 506 516 534 560 562 564 565 566 568 591

WIND: 87 117 126 139 146 149 182 195 283 292 295 305 308 350 367 385 386 387 416
 417 432 434 459 461 498 510 516 560 561 562 564 565 566 567 568 569 573 575 582

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