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Authors
Coles, D.
Concus, Paul
Finn, R.

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D. Coles, P. Concus and R. Finn

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SURFACE TENSION PHENOMENA UNDER LOW- AND ZERO-GRAVITY CONDITIONS I.

by

D. Coles
Graduate Aeronautical Laboratories
California Institute of Technology
Pasadena, California 91109

P. Concus
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

R. Finn
Mathematics Department
Stanford University
Stanford, California 94305

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SURFACE TENSION PHENOMENA UNDER LOW- AND ZERO-GRAVITY CONDITIONS I.

1. Introduction

We report here on the first portion of our study concerning low-gravity capillary free-surface phenomena and related possible Spacelab experiments. The study has three components, mathematical, computational, and experimental, and has been focused primarily on equilibrium capillary surfaces in cylindrical containers (of general cross-section) in the absence of gravity. Central questions are whether or not such surfaces can exist, what their properties are, and what experimental means can be used to observe and to measure them quantitatively.

This study is a continuation of some of our earlier work, which has led to ground-based low-gravity experiments that have been performed in drop towers and in conventional laboratories during the past several years. Although these ground-based experiments have been scientifically productive -- in some ways strikingly so -- they were necessarily limited as to duration of the low-gravity period and controllability of environmental conditions. Some important qualitative features of behavior were observed, but a number of questions were left unanswered. Our recent work has led strongly into areas that require long-term experiments in a carefully controlled low-gravity environment, in order to resolve mathematical uncertainties and provide a precise understanding of the physical phenomena.

2. Existence of menisci in capillary tubes

Our study has centered on equilibrium liquid-vapor free surfaces in cylindrical containers in the absence of gravity, as determined by the
equations

\[ \text{(1)} \quad \text{div } T_u = \text{const. in } \Omega \]

\[ \text{(2)} \quad T_u \cdot \nu = \cos \gamma \quad \text{on } \Sigma, \]

where

\[ T_u = \frac{\nabla u}{(1 + |\nabla u|^2)^{1/2}}. \]

Here, \( \Omega \) is a section of the cylindrical container, \( \Sigma \) is the boundary of \( \Omega \), \( \nu \) is the exterior normal, and \( u(x,y) \) is the (single-valued) height of the surface above a point \((x,y)\) on a reference section. The quantity \( \gamma \) is the contact angle, assumed constant.

In our earlier work we found conditions under which such equilibrium surfaces cannot exist. The conditions depend in general upon an interrelationship involving the local curvature of \( \Sigma \), some global properties of \( \Omega \), and \( \gamma \). When applied to certain particular cases the conditions predicted a discontinuous dependence of the solution on the boundary data (see § 4); this behavior was later verified experimentally at the NASA Lewis Research Center Zero-gravity Facility. For example, for a cylinder with hexagonal cross-section an equilibrium meniscus (solution of (1),(2)) will exist for wetting liquids with contact angles \( \pi/6 \leq \gamma \leq \pi/2 \) (in this case the meniscus is simply a spherical cap); however, if \( 0 \leq \gamma < \pi/6 \), then no solution \( u(x,y) \) of (1),(2) is possible. Figure 9 depicts some NASA Lewis drop-tower experiments illustrating this phenomenon. For case (b) the fluid presumably rises to the top of the container, filling in edges and corners.
Figure 9. Equilibrium free surface at zero-gravity in a hexagonal cylinder. (a) $\gamma = 48^\circ$, (b) $\gamma = 25^\circ$. 
More recently we have found a new set of conditions for existence and for non-existence, which apply to very general cross-sections. For the above case of the hexagonal cylinder, the height at a vertex can be shown to change discontinuously at the critical contact angle. For more general sections, such as the trapezoid discussed in § 4, there remain unresolved important details of the behavior of the capillary surface as the critical configuration is traversed. For example, for a rectangular cross section with a side ratio of 12.5 to 1 our new condition implies the critical angle is $\gamma = \pi/4$ (as it would be for a square); but if the cross section is altered by reducing the length of one of the shorter sides corresponding to a change in the interior angles of only $0.8^\circ$, as in § 4, then no free surface $u(x,y)$ can exist for contact angles even as large as $57^\circ$. As the critical configuration is traversed, there is the question of whether or not the solution surface is bounded. For the trapezoid phenomenon the breakdown in existence apparently occurs for different reasons than for the hexagonal or rectangular cylinders and may not involve a discontinuous change in surface height. This subject is discussed in more detail in § 4.

To investigate the trapezoid and other phenomena experimentally we believe that an in-space experiment is necessary. Because of the much greater sensitivity to boundary perturbations, controlled conditions are required that are not possible to achieve in a drop-tower or other ground-based experiment. Spacelab has promise of providing a suitable environment for exploring these phenomena.
3. **Capillary surface experiments**

Experiments to investigate equilibrium capillary surface properties would involve basically the measurement of the position of surfaces of varying complexity in a number of configurations. For example, for the trapezoid phenomenon the zero-gravity equilibrium surface in cylindrical containers would need to be determined for configurations surrounding the critical one. The height as a function of position would have to be measured, allowing for the possibility that the height may be multiple valued. The photographic recording of position, such as in Fig. 9, would in general not be adequate, since the transition from configurations of existence to non-existence may be complex and require precise determination of the surfaces over most of the cross section. Photographing surface projections on the cylinder walls, as in Fig. 9, can indicate gross qualitative behavior of the surface, but is only suggestive of the actual capillary surface over the interior. Most likely advanced optical methods will be required.

During the completion of our justification study we may find other configurations suitable for Spacelab experiments. One such possible configuration is discussed in § 5. Another would involve the behavior of capillary surfaces under a controlled low- (but non-zero) gravity environment, which might be achieved through rotation of the experiment.
4. Progress in first year's study

4.1. In our first year of study progress has been made toward characterizing mathematically the geometrical conditions on a cylindrical container of general cross section, which determine whether there can be a capillary surface in the container making prescribed angle $\gamma$, $0 \leq \gamma \leq \pi/2$, with the bounding walls. To this purpose, the following result concerning (1),(2) has been proved:

A capillary surface will exist in $\Omega$ with boundary angle $\gamma$, if and only if there is a vector field $\mathbf{y}(x,y)$ in $\Omega$, such that $\text{div} \mathbf{y} = \frac{\Sigma}{\Omega}$ in $\Omega$, $\mathbf{y} \cdot \mathbf{n} = 1$ on $\Sigma$, and $\max_{\Omega \cup \Sigma} |\mathbf{y}| \leq \sec \gamma$.

To prove the result, consider any subdomain $\Omega^*$ as in figure 1. If a vector field of the indicated type exists, we integrate $\text{div} \mathbf{y}$ over $\Omega^*$ to obtain

$$\left| \frac{\Omega^*}{\Omega} - \frac{\Sigma^*}{\Sigma} \right| \cos \gamma < \frac{\Gamma}{\Sigma}.$$  

This is exactly the condition shown by Concus and Finn [1] to be necessary for the existence of a solution, and later by Giusti [2] to be also sufficient. Thus, whenever $\mathbf{y}(x)$ exists, so does a capillary surface.

Conversely, if a capillary surface $u(x,y)$ exists, then the vector field $\mathbf{y}(x) = \frac{\sec \gamma}{\sqrt{1+|\nabla u|^2}} \nabla u$ has exactly the required properties.
The question of finding a capillary surface in a container with section \( \Omega \) is thus reduced to that of finding a vector field \( \psi(x) \) with the specified properties. (Nonexistence can manifest itself physically in various ways, some of which have already been observed experimentally.)

For a triangle or parallelogram, the desired vector field can be constructed explicitly when it exists. For example, for a parallelogram with sides of length 2m inclined with inclination \( \lambda \) to horizontal sides of length 2\( \ell \), we may choose the origin of coordinates to be the intersection point of the diagonals, and set \( \psi = (u,v) \) with

\[
\begin{align*}
u &= \sqrt{1+\lambda^2} \frac{1}{\lambda \ell} x + \left( \frac{1}{m} - \frac{1}{\ell} \right) \sqrt{1+\lambda^2} y \\
v &= \sqrt{1+\lambda^2} \frac{1}{\lambda m} y
\end{align*}
\]

and conclude there is a solution whenever the smaller interior angle \( 2\alpha \) satisfies \( \alpha + \gamma \geq \pi/2 \). If \( \alpha + \gamma < \pi/2 \), we observe from dimensional considerations that in the indicated neighborhood of the angle \( 2\alpha \) (figure 2), there holds

\[
\left| \frac{\Omega^*}{\Omega} - \frac{\Sigma^*}{\Sigma} \right| \cos \gamma > \frac{\Gamma}{\Sigma}
\]

whenever \( \Gamma \) is close enough to the vertex, hence by our earlier results there can be no solution. Since this condition is essentially local in nature, we shall refer to it as the local angle condition. The surface exists whenever \( \alpha + \gamma \geq \pi/2 \),

Figure 2
then it disappears discontinuously if either \( \alpha \) or \( \gamma \) is made smaller.

We obtain a similar result for a section \( \Omega \) as shown in figure 3. Again, there is a solution exactly in those configurations for which \( \alpha + \gamma \geq \pi/2 \), regardless of the side ratios.

![Figure 3](image)

Applying the parallelogram local-angle-condition result to a rectangle, we find there is a solution exactly when \( \gamma \geq \pi/4 \), again regardless of the side ratios.

One might thus expect a similar criterion to hold for a trapezoid. Consider however a trapezoid of base lengths \( a \) and \( b > a \), and smaller vertex angle \( 2\alpha \), as shown. Choosing for \( \Gamma \) the segment half-way up and applying the divergence theorem to \( \Omega^* \), we obtain after some manipulation

\[
\cos \gamma < \frac{2(b+a)^2}{|b-a| + (b+a)\cos 2\alpha} \frac{\cos 2\alpha}{(b-a)}
\]

whenever a solution surface exists. From this relation one sees easily that for any \( \gamma \) in \( 0 \leq \gamma < \pi/2 \), one can choose \( \alpha \) as close to \( \pi/4 \) as desired, and then choose \( a \) and \( b \) in such a way that there will be no
solution. Thus, solutions now fail to exist in configurations that are well within the range for which the local angle condition $\alpha + \gamma \geq \pi/2$ is satisfied.

The situation is at least partly clarified by the result that if the two non-parallel sides of the trapezoid are extended to form with each other an acute angle $2\alpha$, then there exists a solution in the trapezoid whenever $\frac{\pi}{2} - \alpha < \gamma < \pi/2$. Thus, the original local angle condition again enters, in an extended sense. In this case the solution can, however, continue to exist also for smaller values of $\gamma$.

4.2. The question of what happens as a configuration is continuously changed from one in which a solution exists to one in which there is no solution seems of considerable interest. For the local angle phenomenon of the triangle and parallelogram the surface changes discontinuously. To shed light on what happens in the case of the trapezoid (figure 4) we have carried out some numerical computations for particular cases.

It can be shown that the sharpest result corresponding to (3) can be obtained by using for $\Gamma$ a circular arc, instead of the horizontal line shown in figure 4. There will occur $|\nabla u| = \infty$ everywhere on the circular arc in the extremal situation. Thus the breakdown in existence apparently occurs differently in this case than for the case of transition from $\alpha + \gamma \geq \pi/2$ to $\alpha + \gamma < \pi/2$ in the neighborhood of a vertex, such as that depicted in figure 2.

A particular case that we investigated numerically is the trapezoid for which $a = 1.3$, $b = 2$, and the altitude is 25, for which $2\alpha = 89.2^\circ$. For this case it was found that $\gamma_c$, the extremal value of $\gamma$, occurs in (3)
for the circular arc $\Gamma$ of radius $\frac{\Omega}{\Sigma \cos \gamma} = 1.444$ intersecting the slant edges of the trapezoid a distance 7.62 along the slant edges from the top. Thus one expects a capillary surface to exist in the trapezoid (for wetting liquids) for contact angles $\gamma$ greater than $\gamma_c = 57.6^\circ$ but not for contact angles $\gamma$ less than $57.6^\circ$. Note that the local condition $\alpha + \gamma \geq \pi/2$ at the base angle vertex would give information in this case only that a capillary surface would not exist for contact angles $\gamma$ less than $45.4^\circ$.

To investigate the behavior of the capillary surface as the critical condition is approached, (1) and (2) were solved numerically for a sequence of values of $\alpha$ and of $\gamma$. A finite element method was used with reduced biquadratic polynomial basis functions. Because of symmetry, the problem was solved only on the half trapezoid $0 \leq x \leq 1 - 0.014y$, $0 \leq y \leq 25$, with boundary condition $\partial u/\partial x = 0$ on $x = 0$. Both $4 \times 50$ and $7 \times 75$ meshes were used, with good consistency between their results.

The results are depicted in figures 5-7, normalized by the addition of a constant so that $u(0,0) = 10$. In figure 5 the surface height $u(0,y)$ along the symmetry line is shown for several contact angles $\gamma$. The behavior of the solution along other mesh lines in the "y direction" differs very little from that along $u(0,y)$.

In figure 6, the variation of the surface height with $x$ is depicted for several values of $y$ for the case $\gamma = 58^\circ$. Note that the optimal curve $\Gamma$, along which the solution surface would become vertical for the critical contact angle $57.6^\circ$, is a circular arc of radius 1.444 intersecting the symmetry line $x = 0$ of the trapezoid at $y = 17.6$ and the slant edge at $y = 17.4$. 
Figure 5

$u(0,y)$ vs. $y$ for contact angles $75^\circ$, $60^\circ$, $59^\circ$
Figure 6

$u(x, y)$ vs. $x$ for $y = 0, 9, 15, 17.5, 20, 25$; $\gamma = 58^\circ$
Figure 7

$u(0,y)$ vs. $y$ for $a = 2, 1.5, 1.4, 1.3$; $\gamma = 58^\circ$
In figure 7 are depicted the surface heights $u(0,y)$ for a sequence of trapezoids ranging from the rectangle ($a = 2$) to the almost-critical one ($a = 1.3$), for $\gamma = 58^\circ$. The tendency toward verticality is noticeable as criticality is approached.

4.3. The main objective of our current experimental research is to develop optical methods for measuring the position of a capillary surface. The research at present has two components.

One component is to find a working fluid that has satisfactory properties with respect to: (1) surface tension, (2) contact angle, (3) index of refraction, (4) toxicity, (5) flammability, and (6) viscosity. Item (2) interacts with the choice of the container material, as does item (3) if the material is transparent. Information on the listed properties is now being assembled for various fluids.

The other component concerns the optical methods themselves. Optical methods being considered include reflection, refraction, schlieren, shadowgraph, and interferometry (direct or holographic). Recording may be photographic or electronic and in the latter case may involve discrete detectors or closed-circuit television.

Under the present contract, work on finding a satisfactory fluid did not begin until about April, when a suitable student was found. Work on the optical methods is in the conceptual stage and is concentrating on estimates of the relative difficulty and sensitivity of various optical methods. An important element in any decision will be the existing capability of the NASA Fluids Experiment System (FES; see NASA Announcement of Opportunity No. OSTA 80-1) to provide schlieren, holographic, and (with some lens modifications)
shadowgraph records. The volume of the test cell of the FES is adequate for present purposes. Experiments using one or two of the more attractive optical methods will be carried out during the coming summer as part of the first portion of our study. Work will also be begun on techniques for measuring the dependence of contact angle on physical variables such as surface treatment and fluid modification by additives.

5. Proposed second year's study

We propose that our study continue. The trapezoid configuration provides a well-defined basis for examining the feasibility of an in-space experiment, and mathematical, computational, and experimental work can proceed in this direction. In addition, justification of related configurations can be investigated simultaneously.

An example of continued interest is the case of a cylinder cross section consisting of two intersection circles \( \Sigma_1, \Sigma_2 \). Our mathematical results indicate that if the respective radii \( R_1, R_2 \) are equal, then there is always a solution of the capillary equation for any \( \gamma \) in \( 0 \leq \gamma \leq \pi/2 \), regardless of the size of the "opening" between the circles.

However, if the radii are unequal, and if the opening is sufficiently small, then a solution will fail to exist.

The seemingly anomolous behavior is apparently not caused by the infinite curvature at the intersection points of the circles. Whenever \( R_1 \neq R_2 \), it is evidently possible to join the circles by a "neck" region in which the curvatures are as small as desired, as shown in figure 8, but is nevertheless such that no solution will exist.
We propose to continue investigating the "two-circle" configuration and to consider non-zero gravity problems as adjuncts to completing the analytical and numerical study of the trapezoid. After the summer's experimental results are available, we propose to begin planning the substantially expanded experimental work that will be required in the subsequent phases of our study, with [3] as a useful point of departure. For this purpose we propose to be joined by A. Hesselink, an associate of D. Coles at Caltech and authority on optical experimental methods suitable for our experiments. Our results will be embodied in a report that contains a description of the plans developed for the next phase of our study.
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