

Equilibrium Behavior of Fluids in Containers at Zero Gravity

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It is predicted that a wall-wetting fluid in a zero-gravity environment will distribute itself about the tank walls with the vapor centrally located. All liquid-vapor interface areas will have uniform escaping tendency, and therefore the curvature of the bulk liquid surface will be uniform, and the film wet surfaces will assume a thickness consistent with the established escaping tendency. Thus tubes of diameter smaller than the tank diameter will fill with fluid, and the fluid may be collected in desired volumes in the tank by use of baffles.

Nomenclature

A_d	= area of drop of liquid
A_t	= area of tank
A_b	= area of bubble in liquid
ΔF	= free energy
h	= film thickness
k	= constant
n	= thickness in monolayers
P	= vapor pressure over given surface
P_0	= vapor pressure over a flat surface
P_1, P_2	= pressures on each side of a curved surface; P_1 is the pressure in the liquid, P_2 the pressure in the vapor
R	= gas constant
r_1, r_2	= orthogonal radii of curvature. These radii are defined so as to be positive if they lie in the liquid phase (in a liquid drop) and negative in the gas phase (in a vapor bubble)
S	= spreading coefficient
s	= constant
T	= temperature
V	= molar volume
γ_{SV}	= interface free energy between solid and vapor
γ_{LS}	= interface free energy between liquid and solid
γ_{LV}	= interface free energy between liquid and vapor
γ	= interface force = surface tension
θ_e	= equilibrium contact angle

Introduction

THERE are many engineering problems associated with the handling of fluids in zero-gravity fields. Cryogenic liquids are subject to boiloff, and a vent system must be provided which does not expel liquid. Also, fuel lines must be filled before rocket restarts can be made. Knowledge of the spatial configuration of the fluids is required for solutions to many of these problems. The configuration at any time is a function of many things, including interface energies, tank shape, liquid-to-volume ratio, heat transfer, mechanical disturbances, etc. In low earth orbits, drag forces are sufficient to have a large effect on the configuration.

To put these forces in perspective, the rough order of magnitude of the orienting forces present on an orbiting fluid in a tank are given in Table 1.

Heat input is not listed since it cannot be listed properly as a force. The table could be recompiled on a stabilization energy basis, wherein the energies evolved in going from some given configuration (say the rest configuration on the ground) to the stable configuration in orbit are listed. The rough order of magnitude of capillary energy for the model

considered on this basis is 3×10^6 erg. The solar radiant heat incident upon the vehicle is 4×10^{11} erg/sec. Even if 90% of this energy is reflected, energy absorbed from the sun in 1 sec is a million times greater than the capillary stabilization energy. This heat input would lead to thermal gradients that certainly would influence the configuration.

Various treatments of the problems have appeared in the literature. Zero- g fluid handling as it relates to the Agena vehicle has been discussed by Satterlee.¹ The reasonable conclusion is reached that, where drag predominates, the propellants should gather at the forward end of the tank with highly curved gas-liquid surfaces. At higher altitudes, centripetal acceleration forces the fluids to opposite ends of the vehicle. The concept of constant curvature of the liquid-gas interface at equilibrium is mentioned. Mean curvature erroneously is defined as mean temperature.

In discussing a large number of zero- g space powerplant problems, Unterberg and Congelliere² conclude that, if the center of mass is below the propellant tanks, liquids will migrate toward the center of mass during long zero- g coast periods. It is felt that mass attractive forces will exert an influence but that they will be essentially negligible in comparison to other forces present, as shown in Table 1.

A good, detailed treatment of intermolecular forces that determine the behavior of a liquid in a weightless or near-weightless condition has been given by Benedikt.³

In this paper the equilibrium configuration of a fluid with given interfacial free-energy relations, in the absence of a gravity field, is considered. The absence of unbalanced forces, mechanical or thermal, is assumed (except for a qualitative note on the effects of temperature gradients). This is not to imply that the unbalanced forces are not important; indeed, as just noted, such forces may play a dominant role. The determination of the equilibrium configuration is important because it may be used as a starting point for considerations of the thermal and inertial perturbations.

The method employed in this study will be based on interface energy considerations. A similar approach has been taken by Li.⁴ His arguments in establishing that the fluid coats the wall are not rigorous, however, since certain terms have been omitted in the surface free-energy summation equation [namely, the terms $\gamma_{LV}A_b - \gamma_{LV}A_d$ in Eq. (5)].

It should be kept in mind that the establishment of the equilibrium spatial configuration by surface tension manipulation of fluids requires time. If a fluid is displaced from equilibrium, the surface tension forces will tend to form the fluid into the equilibrium spatial configuration, but an "overshoot" may occur. The equilibrium configuration cannot be established until the surface free energy in excess of that energy existing at equilibrium is dissipated. The viscosity of the liquid will cause the excess surface free energy to be converted into heat. The process is, of course, not instantaneous but requires time.

If models are used to study the damping process, it should

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Table 1 Orienting forces on fluid in orbiting tank; rough order of magnitude^a

Gravitational attractive force between liquid mass and vehicle	0.05 dyne
Inertial force due to "aerodynamic" drag, 300-naut-mile orbit	100.00 dynes
Capillary force	1,300.00 dynes
Inertial force due to "aerodynamic" drag, 90-naut-mile orbit	80,000.00 dynes

^a A sphere of liquid hydrogen of 100-cm radius (280 kg) was assumed to be in orbit in a tank of 28 m² cross-sectional area. The center of mass of the hydrogen and the center of mass of the tank plus engines (220 kg) were assumed to be 300 cm apart.

be kept in mind that the time for the attainment of equilibrium will be a function of the size of the container. The mass of fluid to be placed in the equilibrium position increases as the cube of the tank radius. Surface free energy to manipulate this fluid increases only as the square of the radius. Thus, as the tank gets larger, the relative force per unit mass to manipulate the fluid becomes less, and undoubtedly the time required will be longer.

Energy Considerations

Consider a tank containing only a fluid and its vapor. Heat transfer and mechanical disturbances are absent, and the tank and its contents are in equilibrium. There is no gravitational field acting upon the tank. Under these circumstances, the interfacial free energies between the two phases and the material of the tank walls will determine the configuration.

The wetting of a solid by a liquid is governed by the spreading coefficient:^{5, 6}

$$S = \gamma_{SV} - \gamma_{SL} - \gamma_{LV} \\ = -\Delta F_{\text{spreading}} \quad (1)$$

The spreading coefficient is by definition the negative of the free energy change for the process of spreading the liquid over the solid; S may be positive (corresponding to spontaneous spreading) or negative. If, and only if, S is negative or zero, Young's equation holds⁶ (see Fig. 1):

$$\gamma_{SV} = \gamma_{SL} + \gamma_{LV} \cos\theta_e \quad (2)$$

It is found, in general, that on hard solids (which have relatively high surface free energies) most liquids form zero contact angles, θ , and hence, for these systems,

$$\gamma_{SV} \geq \gamma_{SL} + \gamma_{LV} \quad (3)$$

This observation is in accord with current theories of spreading.^{7, 8} Accordingly, this study will be restricted to "wall-wetting" fluids, i.e., fluids that form zero contact angles on the solids that form the tank walls.

From Eq. (3), it may be shown that the fluid in the tank will wet the tank wall and the vapor will accumulate centrally in the tank. Considering a tank that contains a centrally located drop of liquid, the drop is assumed to go from its central location to a configuration in which it is distributed about the sides of the tank, covering all parts of the inner tank surface as shown in Fig. 2. To be as general as possible, it is assumed that the initial drop will have a convex surface throughout, i.e., both radii of curvature will have the same sign. (Actually, the drop will be contained in the form of a

[†] Fluids such as those used for cryogenic propellants have, in general, zero contact angles on steel, aluminum, titanium, and all other metals that have been investigated, and on ceramic or inorganic glassy solids. This behavior, for liquid oxygen, is observed every day in the laboratory or field. For liquid hydrogen, it has been verified in this laboratory by one of the authors; zero contact angles were observed on all solids that were tested.

sphere.) The shape of the tank is arbitrary. It is assumed that the inside area of the tank is greater than the area of a liquid-vapor bubble surface with the fluid distributed about the wall:

$$A_t > A_b \quad (4)$$

This would be true unless A_b had ripples or irregularities that increased its area. At equilibrium, the surface tensions of fluids are such that these irregularities would not occur. The layer covering the walls is of macroscopic thickness so that the liquid-gas interface is not influenced by the underlying solid. The change in the interfacial free energy involved in transition from stage a to stage b is then as follows:

$$\Delta F = -\gamma_{SV}A_t + \gamma_{LS}A_t + \gamma_{LV}(A_b - A_d) \quad (5)$$

If the sum of the terms on the right-hand side of this equation is negative, then the wall wet configuration is favored. This equation may be rearranged to give

$$\Delta F + \gamma_{SV}A_t - \gamma_{LS}A_t = \gamma_{LV}(A_b - A_d) \quad (5a)$$

From Eq. (4) it is obvious that

$$A_t > A_b - A_d \quad (6)$$

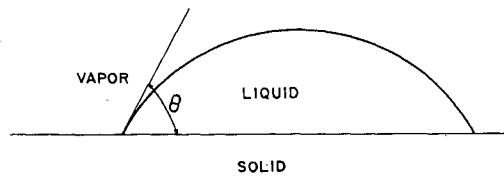
Substituting in Eq. (5a)

$$\Delta F + \gamma_{SV}A_t - \gamma_{LS}A_t < \gamma_{LV}A_t \quad (7)$$

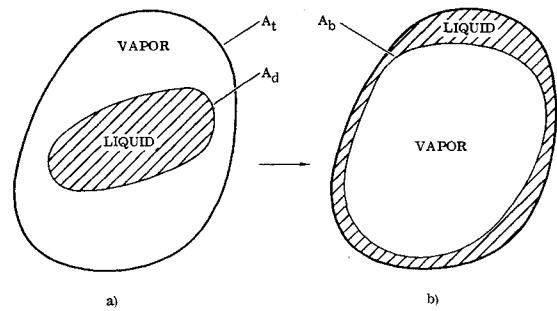
and

$$\Delta F < A_t(-\gamma_{SV} + \gamma_{LS} + \gamma_{LV}) \quad (7a)$$

Now referring to Eq. (3), if the equal sign is assumed, the quantity in parentheses in Eq. (7a) is equal to zero, and, accordingly, the free energy is less than zero or negative. If the inequality is assumed, the free energy is less than the negative quantity and is negative. Thus the wet-wall configuration is favored over the centrally located drop. Up to this point the exact configuration of the wet wall has not been stated except to note that it does not contain ripples, etc.



$$\gamma_{SV} \quad \gamma_{LV} \quad \theta \quad \gamma_{LS}$$

Fig. 1 Contact angle

A_t = AREA OF THE INNER TANK SURFACE

A_d = AREA OF THE SURFACE OF THE DROP

A_b = AREA OF THE INNER BUBBLE SURFACE

Fig. 2 Schematic of change from central liquid drop to wall-wet surfaces with central vapor bubble

Consideration now will be given to the wet-wall configuration that is the most stable, i.e., has the lowest free energy.

The configuration of the centrally located vapor pocket at equilibrium will be one of minimum interfacial free energy. Since only the liquid-vapor surface is a variable, the problem becomes one of minimizing the surface free energy for a given vapor volume. If the tank configuration is such that the vapor volume can form a sphere, it will form and will be the stable configuration, as discussed in Ref. 9. If the sphere that would contain the vapor has a larger diameter than the smallest dimension through the center of the tank, the situation is more complicated.

A cylindrical tank with hemispherical ends which contains a wetting fluid under zero gravity is assumed. The interfacial energies are represented by the inequality (3). Consider the spatial configurations existing at equilibrium as the fluid is removed and replaced by its vapor. A sphere will form and grow in the central portion of the tank. The position along the axis of the cylinder is arbitrary. The bubble eventually will approach the walls of the cylinder as illustrated in parts a, b, and c of Fig. 3. (As noted below, the vapor always will be separated from the wall by an adsorbed film; it is this condition that is referred to as a "film-wet" surface.)

Up to this point the treatment of the problem has been based on interfacial free energy considerations. To proceed past this point and predict the spatial configuration resulting upon the removal of more fluid, surface curvature considerations will be introduced. The curvature approach is consistent with the surface free energy minimization approach. The curvature approach allows (perhaps) an easier means of visualizing the problem.

Surface Curvature: Vapor Pressure Considerations

In general practice, the term "vapor pressure" refers to the pressure of a vapor in equilibrium with a liquid that has a flat surface. The vapor pressure is a measure of the escaping tendency of the liquid. The equilibrium pressure over a curved surface is different from the pressure over the flat surface; a drop of liquid has a higher equilibrium vapor pressure than a flat surface; a vapor bubble in a volume of liquid has a lower equilibrium vapor pressure than a flat surface.⁶ For curvatures in which the radius involved is

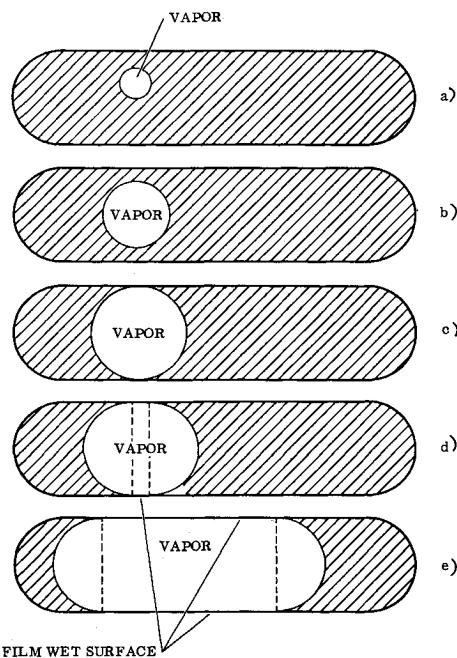
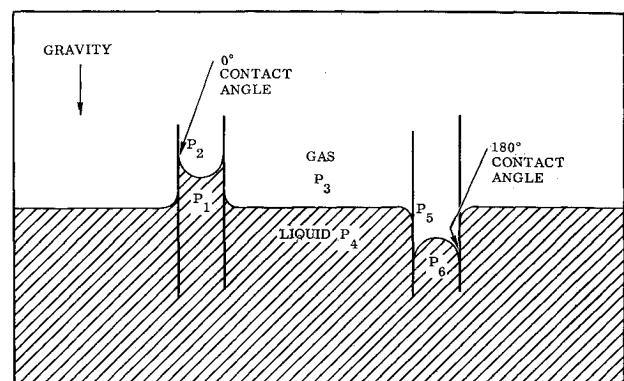


Fig. 3 Vapor bubble; equilibrium conditions, zero-g



P_1, P_4, P_6 = PRESSURES BELOW MEMBRANE

P_2, P_3, P_5 = PRESSURES ABOVE MEMBRANE

Fig. 4 Pressures across capillary membranes

the order of millimeters or centimeters or larger, the differences in pressure are exceedingly small. The relationship between the curvature and the equilibrium pressure is given by Eq. (8):

$$RT \ln(P/P_0) = \gamma V[(1/r_1) + (1/r_2)] \quad (8)$$

To avoid confusion regarding the various "pressures" in a system, a short discussion may be given wherein the various pressures are put in relative order. The equation of capillarity relates the pressures on the two sides of a curved surface:

$$P_1 - P_2 = \gamma [(1/r_1) + (1/r_2)] \quad (9)$$

A drop of liquid will have a higher internal hydrostatic pressure than the gas surrounding it, whereas a vapor bubble will have a higher pressure than the liquid around it. In Eq. (8) the pressures involved are vapor pressures over a flat surface compared to a curved surface.

Consider a liquid in the earth's gravitational field into which two capillaries are projected, one capillary having a surface that is wetted by the liquid (contact angle = 0°) and the other capillary having a different surface that is not wetted by the liquid (contact angle = 180°). Assume only the liquid and its vapor to be present. The fluids will assume configurations as shown in Fig. 4 (see Ref. 6, p. 10). The pressures across the menisci will assume certain values depending on the curvature of the menisci. According to Eq. (9), hydrostatic pressures across the menisci are represented qualitatively as follows:

$$P_6 > P_5 \quad (10)$$

$$P_3 = P_4 \quad (11)$$

$$P_2 > P_1 \quad (12)$$

According to Eq. (8) the relative vapor pressures are as follows:

$$P_5 > P_3 > P_2 \quad (13)$$

Since only the vapor of the liquid is present, the pressure in the gas phase is the same as the vapor pressure. From the two sets of relative pressures, the order of decreasing pressure may be listed as follows:

$$P_6 > P_5 > P_4 (= P_3) > P_2 > P_1 \quad (14)$$

If the vessel were opened to the air, the pressures would increase throughout the system in an almost uniform increment (almost the same increment of pressure increase would be added to all the pressures), and the relative pressure order given in Eq. (14) would remain unchanged.

The system shown in Fig. 4 is in equilibrium, even though the pressure is greater over the right-hand meniscus than over the left-hand meniscus. The pressure existing because of meniscus curvature is consistent with the gravity pressure gradient.

Similar reasoning may be applied to the zero-gravity case. Consider the situations as given in Fig. 5. In Fig. 5a, a bubble of vapor exists in a body of liquid; in Fig. 5b, a flat surface exists in contact with the vapor (rather difficult to obtain experimentally, but easy enough to imagine); and in Fig. 5c, a drop of liquid exists at equilibrium with vapor around it. The system consists of one component in two phases: liquid and vapor. The pressure notation is the same as given in Fig. 4, and the pressure relationships are the same as those given in Eq. (14).

Returning now to Eq. (8), in the problem considered, V , P_0 , and γ are constant. Thus the curvature as represented by $(1/r_1) + (1/r_2)$ determines the vapor pressure over the curved surface. Radii in the liquid are positive, and radii in the vapor are negative. In any situation occurring in a tank, in order that equilibrium be established, it is necessary that the pressure be uniform throughout the vapor volume. It follows that the curvature will be constant for all surfaces at equilibrium.

Returning to the example given in Fig. 3, as the amount of liquid is reduced below the amount shown in Fig. 3c, while equilibrium is maintained in the system, it is predicted that the configuration shown in Fig. 3d (see Ref. 9) will result. The vapor here is contained in a volume that is a cylinder in its central portion, enclosed on both ends by hemispheres. If the radius has a length r' , then the curvature of the hemispherical ends

$$(1/r') + (1/r') = -2/r$$

will impose a particular vapor pressure upon the system. If the cylindrical connecting section of liquid were backed by bulk liquid, the vapor pressure over the cylindrical surface would be greater than the vapor pressure above the spherical surfaces because of the larger total curvature of the surface

$$-1/r' > -2/r'$$

At equilibrium this situation could not exist.

Under these circumstances, it may be predicted that the layer of liquid in the cylindrical portion will evaporate partially and become thinner, until a film thickness is reached such that the liquid-vapor surface is close enough to the solid that the attractive force on surface molecules, due to the solid, is appreciable. This extra attraction, due to the underlying solid, will reduce the vapor pressure over the film until the vapor pressure over the cylindrical portion is equal to that over the hemispherical portion.

As more liquid is removed from the cylinder, the hemispherical bubble ends will move towards the ends of the cylinder, and more and more cylindrical, film-wet surface will be created in the midsection.

Film Thickness

The equilibrium thickness of the film may be predicted from adsorption isotherms. For physical adsorption on a flat surface at high coverage (more than three or four times monolayer coverage), adsorption generally follows the Frenkel-Halsey-Hill equation:¹⁰⁻¹³

$$\ln(P/P_0) = -(k/n^s) \text{ or } n = [-k/\ln(P/P_0)]^{1/s} \quad (15)$$

MacMillan and Teller¹⁴ have shown theoretically that k is of the order of magnitude of unity, and s is approximately 3.

No reliable data are available as to the adsorption of hydrogen or oxygen on steel (other than chemisorption, which is limited to monolayer coverage) or on oxides of iron supported by the metal; but data on the adsorption of oxygen

on anatase (TiO_2) may be used to estimate the order of magnitude of the film thickness. For this system, it has been reported^{12,13} that $s = 3$ and $k = 4$.

Consider a system in which a flat surface covered by an adsorbed film of liquid is in equilibrium with a concave surface of bulk liquid having equal radii of curvature. For this system, Eqs. (8) and (15) may be combined as follows:

$$-k/n^s = \gamma(V/RT)[(1/r_1) + (1/r_2)] \quad (16)$$

Since $r_1 = r_2$

$$n = (-kRTr/2\gamma V)^{1/s} \quad (17)$$

When the solid surface on which the adsorption takes place is strongly concave, the adsorbed film will be thicker than that given by Eq. (17). (This is because, by the derivation of Eq. (15), there will be more of the solid close enough to a given molecule in the film to exert an appreciable attractive force on it if the surface is concave than if it is flat. If the surface is highly convex, then the film will be thinner than on a flat surface.) However, for surfaces such as those involved in cryogenic rocket tankage, the radii of curvature are, in general, large enough that the surface can be considered to be flat for this calculation.

For oxygen at 90°K, $\gamma = 13.2 \text{ dyne/cm}$, and $V = 28.1 \text{ cm}^3/\text{mole}$. The thickness of a monomolecular layer of O_2 is about 4 Å; so if h is the film thickness in centimeters, $h = 4 \times 10^{-8}n$. For a tank 10 ft in diameter, and with the gas bubble as indicated in Fig. 3d or 3e, the radius r is 150 cm. Hence

$$\begin{aligned} h &= 4 \times 10^{-8}(2RTr/\gamma V)^{1/3} \\ &= 7.5 \times 10^{-5} \text{ cm} = 750 \text{ m}\mu \end{aligned}$$

For liquid hydrogen at 20°K, the film thickness should be of the same magnitude as for oxygen at 90°K, namely, about 750 m μ and in any case much less than 1 mm thick. The accuracy of these estimates is limited; however, the film thickness is clearly very small.

A film such as this is thick enough that an experimental test of the prediction might be considered; an interferometric method, e.g., an ellipsometer, easily could measure the film thickness. But such experiments are actually out of the question, for another reason.

For liquid oxygen at its boiling point, $dP/dT \cong 77 \text{ mm}/^{\circ}\text{C}$. For liquid hydrogen at its boiling point, $dP/dT \cong 200 \text{ mm}/^{\circ}\text{C}$. From Eq. (8) the reduction in vapor pressure due to negative curvature for a 150-cm radius tank is

$$(P - P_0)/P_0 \cong 6.7 \times 10^{-10}$$

i.e., the vapor pressure is only about $5 \times 10^{-7} \text{ mm Hg}$ below the saturation pressure. So, to carry out the experimental test, it would be necessary to "thermostat" the tank to within about $\pm 10^{-8} \text{ }^{\circ}\text{K}$ in the case of oxygen or $10^{-9} \text{ }^{\circ}\text{K}$ in the case of hydrogen.

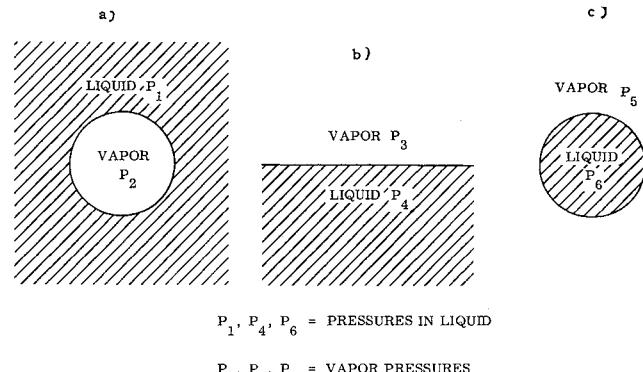


Fig. 5 Pressures across capillary membranes; zero-g

Effect of Temperature Gradients

For hydrogen at 20°K, $d\gamma/dT \cong -0.2$ dyne-cm⁻¹·deg⁻¹. For oxygen at 90°K, $d\gamma/dT \cong -0.25$ dyne-cm⁻¹·deg⁻¹. If there were a higher rate of heat flow through one wall of a tank than through the other walls, so that the bubble surface became warmer on one side than the other, then the surface tension would be reduced over the warm part of the bubble. This reduction in surface tension would allow the warm surface to be "pulled" towards the colder surface areas. The gross effect on the bubble would be a movement in the direction of the source of heat. In any case, the overall result would be a different spatial configuration of the fluid. The calculation of the actual distribution of fluids in a nonisothermal system at zero gravity would be extremely difficult, even for very simple configurations of tanks, walls, and heat sources. There might not be a "stationary," steady-state configuration, but, rather, oscillations even might result.

Predictions

On the basis of the arguments made, predictions may be made for a wall-wetting fluid and its vapor, in equilibrium in

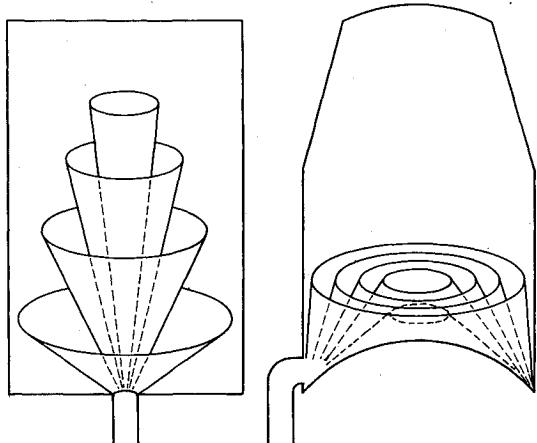


Fig. 6 Zero-g liquid collectors

a tank, under a zero-gravity field, and in the absence of mechanical and thermal disturbances:

- 1) The fluid will wet the wall and distribute itself so as to form a vapor space in the central portion of the tank.
- 2) If a sphere that contains the vapor space has a diameter less than the smallest diameter of a vessel, then this sphere, located away from the wall in the tank (or in the extreme, just barely tangent to the wall), will represent the stable configuration.
- 3) If the sphere containing the vapor volume has a diameter greater than the small dimension of the tank, a centrally located vapor space will result with the liquid distributed so that the surfaces backed by bulk liquid are of uniform curvature; other surfaces will be film-wetted tank walls. The escaping tendency from the wetted tank walls will be adjusted by evaporation (and thus reduction in thickness) to equal the escaping tendency from the curved surfaces. Thus, the escaping tendency of the surfaces exposed to the vapor volume will be uniform.

4) The curvature of a bulk liquid surface will be greater than or equal to the curvature of any exposed film-wet surface. (Otherwise, the liquid would collect on the film-wet surface.)

For similar systems displaced from equilibrium, predictions are made as follows:

- 1) Mass rearrangement to give equilibrium may occur by flow in the liquid phase and by evaporation from an area of

high escaping tendency and condensation in an area of low escaping tendency. (In the case of the movement through the vapor phase, the inequality in vapor pressure rapidly translates itself into an inequality in temperature, since the surface from which the fluid is evaporating becomes cooled and the surface upon which the condensation is occurring becomes heated. The rate of mass transfer then is controlled by the heat transfer. Under zero-gravity conditions heat transfer occurs mainly by conduction, since convection is not present, and, with small temperature differentials at low temperatures such as 20°K, radiative heat transfer is rather slow. If the tank wall is a good conductor or if there are other members in the tank which are good conductors, the rate of heat transfer will, of course, be accelerated.)

2) If a steady source of heat is present which maintains one region of the walls appreciably warmer than other regions, then one area of the vapor bubble would become warm, and this warm film would be pulled to the colder surface. If other phenomena are not active to replenish the fluid in the warm area, eventually the warm tank surface should be free from bulk liquid and merely "film-wet." Evaporation and condensation would tend to produce the same result.

From these rules certain conclusions regarding space-vehicle hardware may be drawn. Tubes with large curvature, i.e., small diameter, will fill with fluid, and the surface at the opening of the tube will assume a shape consistent with the vapor pressure in the tank. Thus a vapor vent tube that relies for its performance on its projecting into the vapor space should have the valve on the end of the tube in the vapor space. Fuel lines leading from the tank, if opened to the tank, should fill with fluid (providing fuel line temperature phenomena are not over-riding).

By properly shaping the surfaces, it would appear to be possible, given sufficient time, to cause fluid to collect in any volume desired. For example, a tank of conical shape would collect the fluid at the apex end. If it were desired to design a tank employing this principle, a honeycomb of cones (or "hexagonal cones") with a common apex at the exit port of the tank could be used. This would provide a "wick" for the fluid. The fluid could be collected centrally (away from walls). If bubbles resulted from heat absorption, these would tend to move away from the apex. Two other arrangements in the same category are shown in Fig. 6.

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