percent methane in the mixture increased, the percent deviation increased. The discrepancy between predicted and experimental values indicates that predictions based on these solid-vapor ratios can only be used as a first approximation. In mixtures of hydrates containing olefinic hydrocarbons the equilibrium ratios for the individual components are evidently a function of composition. This can only be established quantitatively by analysis of hydrates formed from a vapor of fixed composition.

DISCUSSION

Experimental data for the ethylene-water system show that the pressure-temperature gradient of the curve representing the hydrate—vapor—water-rich liquid equilibrium in this system passes through two points of inflection between pressures of about 700 and 1,100 lb./sq. in. abs. The reason for this is not clear. It is known that the number of molecules of water associated with each molecule of gas in the hydrate lattice is different for different systems. It is also known that a change in crystal structure is associated with a change in slope of the vapor-pressure curve. It is suggested that the points of inflection observed for this system result from a change in the number of water molecules associated with the hydrocarbon molecule causing a change in the crystal structure of the hydrate.

In methane-ethylene-water systems containing higher concentrations of ethylene the appearance of two inflections similar to those for the ethylene-water system is observed. At lower concentrations the curves are similar and essentially parallel to the curve for the methane-water system. This is to be expected because each of these curves is a locus of points at fixed composition on a surface in space connecting the pure component curves. The folding of this surface appears to be such that at pressures above about 12,000 lb./sq. in. abs. ethylene hydrates will form at temperatures lower than for methane hydrates.

An interesting aspect of the configuration of this surface is that constant-pressure sections show a maximum temperature such that hydrates in the mixture of hydrocarbons form at a temperature above that for either single component. To the best of the authors' knowledge the hydrogen sulfide-propane system is the only other system known to exhibit this behavior (*7*).

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Taylor Instability of an Evaporating Plane Interface

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A new mode of high-flux heat transfer is proposed, which consists of film boiling from a porous solid plate, the vapor being sucked off through the plate. For horizontal plates the average vapor film thickness is determined by Taylor instability of the vapor-liquid interface. A theory is presented for predicting the minimum vapor film thickness which takes into account the stabilizing influence of the reactive pressure of the vapor leaving the interface. The predicted heat transfer coefficients are five to ten times greater than observed heat transfer coefficients in film boiling from nonporous surfaces.

Sir Geoffrey Taylor first called attention to the instability which results when an interface between two fluid phases of unequal density is accelerated away from the lighter phase (*1*). This is exactly equivalent to the instability of the interface between a dense fluid lying over a lighter fluid. Such a condition exists in film boiling of a liquid from a horizontal flat plate, where a quasistable film of vapor separates the liquid from the plate. The zone is stabilized by the presence of the hot surface which results in evaporation from the interface. Previous treatments have considered the modifying effects on Taylor instability of surface tension (*2*), viscosity (*2*), finite depth of fluids (*3*), and velocity of the fluids parallel to the interface (combined Taylor and Helmholtz instability) (*3*);

the purpose of this paper is to describe the stabilizing effect of evaporation from the interface.

The problem has some practical applications. Previous analyses (*4, 5*) of *film boiling* from tubes have been based upon the hydrodynamics of vapor flowing from the *point* of generation to the point of collection and removal from the film. Thus the mean film thickness, and hence the heat transfer coefficient, at any point is determined by the vapor flow parallel to the heating surface. It is however possible to conceive of a geometry where the removal of vapor would not be by a flow parallel to the heating surface but rather through the porous heating surface itself. In this case the interface can approach as close to the heating surface as the evaporation from the interface will allow. The mean thickness of the vapor film is now determined by Taylor instability considerations rather than vapor removal. Intuitively one would therefore expect considerably higher transfer coefficients in film boiling from porous horizontal plates than from solid horizontal plates. This expectation is confirmed in the analysis which follows. These results may be of practical significance in the design of nuclear reactors and zero-gravity heat transfer systems.

STATEMENT OF PROBLEM

Consider a liquid of unlimited extent which is at rest at infinity and which lies above a hot solid plate. The temperature T_w of the plate is sufficient so that a film of vapor of mean thickness separates the liquid from the plate. The vapor is removed as fast as it is formed by passing through pores in the solid plate. Assume that the heat conduction into the bulk liquid is negligible, or equivalently that the liquid is everywhere at the saturation temperature. The problem now is to determine the stability of the liquid-vapor interface to small perturbations. The coordinate system is fixed at the mean vapor-liquid interface, the positive direction for y being upward, so that the solid surface is at $y = -b$.

Assume the liquid to have a velocity potential such that

$$\frac{\partial^2 \phi}{\partial x^2} + \frac{\partial^2 \phi}{\partial y^2} = 0; \quad \phi = \phi\,(x, y, t) \quad (1)$$

$$y > 0, t > 0$$

The linearized kinematic condition at the free surface states that

$$\frac{\partial \phi}{\partial y} = -\frac{\partial \eta}{\partial t}; \quad \eta = \eta\,(x, t) \quad (2)$$

This is equivalent to equating the vertical velocity of a free surface at any point to the particle velocity at the point, for small perturbations of the surface.

The pressure equation at the liquid-vapor interface gives

$$\rho_L \frac{\partial \phi}{\partial t} = g'\eta\,(\rho_L - \rho_V) + \sigma \frac{\partial^2 \eta}{\partial x^2} + \rho_V\, v_e^2 \quad (3)$$

Essentially this equation states that the inertial pressure of the liquid at the interface is balanced by the hydrostatic pressure difference, by the pressure difference due to surface tension, and by the reactive pressure of the vapor leaving the liquid surface. In Equation (3) one is concerned only with the variable portion of the reactive pressure, the constant part being absorbed by suitable definition of the velocity potential.

In order to determine the magnitude of the last term in Equation (3) it is necessary to consider the heat flow in the vapor. The heat-flow equation in the vapor, on the assumption of steady one-dimensional flow may be written

$$v_e\, \zeta(y) = \alpha_V \frac{d\,\zeta(y)}{d\,y};$$

$$\zeta(y) \equiv \frac{dT}{dy}, -b < y < \eta \quad (4)$$

Density changes of the vapor in flowing across the film thickness are neglected in this approximation. The heat balance at the liquid-vapor interface requires

$$k_V\, \zeta(\eta) = \Lambda\, \rho_V\, v_e \quad (5)$$

Integrating Equation (4) twice and making use of Equation (5) one obtains

$$v_e = -\frac{\alpha_V}{b + \eta} \ln \left[1 + \frac{C_p(T_w - T_s)}{\Lambda}\right] \quad (6)$$

For small perturbations of the interface therefore

$$\rho_V\, v_e^2 \cong B\left[1 - \frac{2\eta}{b}\right]; \frac{\eta}{b} << 1 \quad (7)$$

where

$$B = \rho_V \left\{\frac{\alpha_V}{b} \ln\left[1 + \frac{C_p(T_w - T_s)}{\Lambda}\right]\right\}^2 \quad (8)$$

Equation (3) therefore becomes upon the substitution of the variable part of the reactive pressure

$$\rho_L \frac{\partial \phi}{\partial t} = g'\eta\,(\rho_L - \rho_V) + \sigma \frac{\partial^2 \eta}{\partial x^2} - \frac{2\,\eta\, B}{b} \quad (9)$$

Equations (1), (2), and (9) constitute a complete statement of the problem. One may investigate the stability of this system by classical methods, whereby one determines whether small harmonic perturbations of the system tend to decay or to grow. This is the procedure employed in references *1* to *3*. However since the thickness of the vapor film may in some cases be a variable with time, it is instructive to consider this problem as an initial-value problem and to determine the behavior of the system for large values of time. This is the procedure used herein.

A preliminary simplification is possible. In virtue of the Fourier integral theorem a single Fourier component in x may be considered, without loss of generality:

$$\phi(x, y, t) = \xi(y, t)\, e^{imx} \quad (10)$$

$$\eta(x, t) = \mu(t)\, e^{imx} \quad (11)$$

where it is understood that the real parts of the complex quantities are to be considered. In terms of these substitutions Equations (1), (2), and (9) become

$$\frac{\partial^2\, \xi(y, t)}{\partial y^2} - m^2\, \xi(y, t) = 0 \quad (12)$$

$$\frac{\partial\, \xi(0, t)}{dy} + \frac{d\,\mu(t)}{dt} = 0 \quad (13)$$

$$-\rho_L \frac{\partial\, \xi(0, t)}{\partial t} + \beta\, \mu(t) = 0 \quad (14)$$

where the space-independent part of the reactive pressure in Equation (3) has not been included, since the addition of a constant pressure everywhere in a system can have no dynamical effect on an incompressible flow. One now assumes that initially the system is everywhere at rest, while the surface is perturbed from its mean position. This may be written as

$$\xi(y, 0) = 0 \quad (15)$$

$$\mu(0) = \mu_o \quad (16)$$

In these equations

$$\beta = g'\,(\rho_L - \rho_V) - \sigma m^2 - \frac{2B}{b} \quad (17)$$

SOLUTION OF INITIAL-VALUE PROBLEM

Taking the Laplace transform of Equations (12) to (14) with respect to time and utilizing the initial conditions (15) and (16) one obtains

$$\frac{\partial^2\, \overline{\xi}(y, s)}{\partial y^2} - \mathrm{m}^2\, \overline{\xi}(y, s) = 0 \quad (18)$$

$$\frac{\partial\, \overline{\xi}(0, s)}{\partial y} + s\, \overline{\mu}(s) - \mu_o = 0 \quad (19)$$

$$-s\, \rho_L\, \overline{\xi}(0, s) + \beta\, \overline{\mu}(s) = 0 \quad (20)$$

where

$$\overline{\xi}(0, s) = \int_0^\infty e^{-st}\, \xi(0, t)\, dt$$
$$\equiv \pounds\ \{\xi(0, t)\} \quad (21)$$

Equation (18) is satisfied by

$$\overline{\xi}(y, s) = \overline{A}(s)\, e^{-my} \quad (22)$$

as the solution which is bounded at infinity. Inserting this result into Equation (19) one obtains

$$\overline{\mu}(s) = \frac{m\,\overline{A}(s) + \mu_o}{s} \quad (23)$$

With these results Equation (20) yields

$$\overline{A}(s) = \frac{\beta\mu_o}{\rho_L\, s^2 - m\beta} \quad (24)$$

and the amplitude of the surface perturbation will be unstable if and only if

$$\beta \geqslant 0 \quad (25)$$

Consideration of the mean vapor film thickness as a parameter implies an upper bound on b in order for the vapor film to be stable:

$$b < \left(\alpha_v \ln\left[1 + \frac{C_p(T_w - T_s)}{\Lambda} \right]\right)^{2/3}$$
$$\left(\frac{2\,\rho_v}{g'(\rho_L - \rho_v) - \sigma m^2} \right)^{1/3} \quad (26)$$

Since the local heat flux is given by

$$q_l = \Lambda\, \rho_v\, v_e \quad (27)$$

this leads to the space-average heat flux

$$q = \Lambda \left(\frac{k_v\, \rho_v}{2\, C_p} \left[g'(\rho_L - \rho_v) - \sigma m^2 \right] \right.$$
$$\left. \ln\left[1 + \frac{C_p(T_w - T_s)}{\Lambda} \right] \right)^{1/3} \quad (28)$$

For moderate wall superheats this may be simplified to read

$$q \sim \left\{ k_v\, \rho_v \frac{\Lambda^2}{2} \left[g'(\rho_L - \rho_v) - \sigma m^2 \right] \right.$$
$$\left. (T_w - T_s) \right\}^{1/3} ; \frac{C_p(T_w - T_s)}{\Lambda} << 1 \quad (29)$$

For the fully stable case where all wave lengths are allowed (infinite flat plate) $m = 0$ in the above expressions. This indicates that the stabilizing effect of surface tension becomes negligibly small for long wave lengths.

Equations (28) and (29) indicate that the limiting heat transfer coefficient in film boiling varies approximately as $(T_w - T_s)^{-2/3}$. Since $\Lambda^2\ \rho_v$ goes through a maximum in the range of reduced pressures from 0.2 to 0.6, h will also go through a maximum in this range.

Experimental data on film boiling from porous plates are not available for comparison with this theory. It is of interest however to note the order of magnitude of the predicted heat transfer coefficient in comparison with those reported for film boiling from nonporous surfaces. Heat transfer coefficients in the range of 25 to 90 B.t.u./(hr.)(sq. ft.)(°F.) have been reported (6) for film boiling of oxygen from a 0.069-in., horizontal, stainless steel tube over a range of pressures of 7.2 to 487 lb./sq. in. abs. For film boiling of pentane and carbon tetrachloride from a horizontal flat plate at atmospheric pressure (7), heat transfer coefficients of 25 to 50 B.t.u./(hr.)(sq. ft.)(°F.) have been reported. By comparison heat transfer coefficients calculated by Equation (29) for film boiling from a horizontal porous plate under similar conditions are predicted to be of the order of five to ten times as great as these results. The predicted heat fluxes may well be high however because of the neglect of the insulating effect of the surface pores on the heat transfer to the vapor-liquid interface. On the other hand brief contacts between the liquid and the solid may occur, induced by turbulent effects, which would increase the average heat transfer coefficient. These questions must be solved by experiment.

In conclusion one may note some advantages for nuclear, space, and other compact heat exchanger applications of this mode of heat transfer over nucleate boiling, which is the conventional high-flux heat transfer mode. Nucleate boiling is dependent upon the functioning of statistical surface roughnesses, whose activation and deactivation upon change in the heat flux rate, liquid flow rate, pressure, or other boiling conditions exhibit appreciable time lags. Hence nucleate boiling systems exhibit undesirable resonances and instabilities in closed-loop systems. Porous-element film boiling, since it is not dependent upon surface nucleation, should be free from this difficulty. Further, since the fuel element temperature is higher than it is in nucleate boiling, one would expect the thermodynamic efficiency to be greater, implying appreciable superheating of the vapor. Since there should be negligible liquid-solid contact, corrosion problems may be minimized. Further, the cost of a porous, ceramic fuel element may well be considerably below that of a metallic clad element.

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NOTATION

$\overline{A}(s)$ = transformed velocity potential time-dependence factor, Equation (22)
B = function defined by Equation (8)
b = mean vapor film thickness
C_p = specific heat of vapor
g' = acceleration due to gravity and all other causes
h = heat transfer coefficient
k_v = thermal conductivity of vapor
m = wave number
q_l = heat flux, local
q = heat flux, space average
s = transform time variable
T_s = saturation temperature
T_w = wall temperature
t = time
v_e = velocity of vapor evaporating from interface
x = distance parallel to interface
y = distance perpendicular to interface

Greek Letters

α = thermal diffusivity of vapor
β = function defined by Equation (17)
ζ = temperature gradient, Equation (4)
η = elevation of interface
Λ = latent heat of vaporization
μ = amplitude of Fourier component of interface elevation
ξ = amplitude of Fourier component of velocity potential
ρ_L = density of liquid
ρ_v = density of vapor
σ = surface tension
ϕ = velocity potential of liquid

Superscripts

— = Laplace transform with respect to time

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