

to be unstable to smaller amplitude disturbances. Further work is needed to identify quantitative limits.

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## The Influence of Lateral Pressure Variations on the Stability of Rapidly Evaporating Liquids at Reduced Pressure

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As the evaporation rate of a liquid under vacuum ( $<1$  torr) is gradually increased, a critical point is reached at which the gas-liquid interface spontaneously transforms from a relatively quiescent surface to one which is intensely turbulent, producing a marked increase in the evaporative flux (Hickman, 1952, 1972). The instability is attributed to the differential vapor-recoil mechanism in which the interface is unstable to local variations in evaporation rate which are produced by local perturbations in surface temperature, local surface depressions being produced by the recoil force exerted on the surface by the rapidly departing vapor and sustained liquid flows being driven by the resultant shear exerted on the liquid surface by the vapor (Palmer, 1976). Interfacial contamination can play a major role in determining the stability of the liquid surface. In particular, both linear stability analysis and experiments have shown that the addition of trace amounts of a nonvolatile surfactant to a liquid of moderate surface tension can dramatically increase the stability limit for vapor-recoil instability (Palmer, 1977).

In addition to establishing criteria for vapor-recoil instability, the linear stability analysis of a rapidly evaporating liquid also revealed that the interface may become unstable owing to lateral variations in gas phase pressure at the interface which are produced when fluid streamlines are distorted near the deforming interface. Called the fluid-inertia mechanism, this mode of instability is unaffected by the degree of surfactant contamination present in the system and therefore establishes an upper bound on the stability of the interface.

Although the analysis of Palmer (1976) accounts for the effect of fluid inertia on pressure fluctuations through the equation of momentum conservation for the gas

phase, it does not consider how these lateral pressure variations influence the local evaporation rate and thus the criteria for instability. The rate of evaporation depends not only on surface temperature but also on gas phase pressure (see Schrage, 1953). In the vicinity of surface depressions, inertial effects produce an increase in gas phase pressure and consequently a decrease in the local evaporation rate. From the viewpoint of vapor-recoil instability, the effect is to moderate the local increase in evaporation rate and recoil force which accompanies local increases in surface temperature. From the viewpoint of fluid-inertia instability (for which the surface temperature is presumed constant), the effect of local pressure variations on evaporative flux is to produce a recoil force which is maximum on surface crests and minimum in depressions, thus resisting further deformation of the interface.

In summary, then, the effect of gas phase pressure on evaporation rate is to increase the stability of the system to disturbances amplified by both the differential vapor-recoil and fluid-inertia mechanisms. The purpose of the current paper is to assess the importance of this stabilizing influence with the aid of linear stability analysis.

#### MATHEMATICAL ANALYSIS

The linear stability analysis of a steadily evaporating liquid in which pressure effects on evaporation rate are considered differs from that presented by Palmer (1976) only in the expression for the perturbed evaporation rate  $\eta'$ , which for the present case is given by

$$\eta' = \left( \frac{\partial \eta}{\partial T} \right) T' + \left( \frac{\partial \eta}{\partial p_v} \right) p_v'$$

The result is that the term  $N_H(T_L - B)$  in Equations (24) and (27) of Palmer (1976) is replaced by  $N_H(T_L - B) - N_{p_v}$ . All the other differential equations and

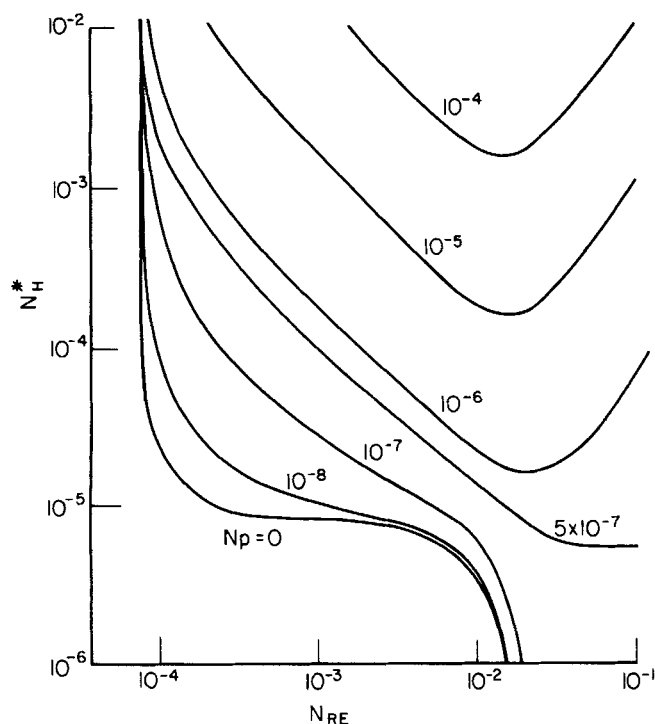


Fig. 1. The effect of pressure number on the relationship between critical Hickman number and liquid phase Reynolds number for conditions  $N_p = 10^8$ ,  $N_{PR} = 10$ ,  $N_\mu = 10^2$ ,  $N_{BO} = 1$ ,  $N_{CR} = 10^{-5}$ .

boundary conditions remain unchanged in the present analysis and for brevity are not repeated here.\*

The dimensionless group  $N_p$  is new to the analysis and reflects the degree to which the stability of the system is increased by the influence of gas phase pressure on local evaporation rate. In particular,  $N_p$  represents the ratio of the stabilizing effect due to the influence of gas phase pressure on the local recoil force to the stabilizing effect of surface tension which resists surface deformations.

To simplify the analysis, the effects of kinetic energy transport and viscous heating at the interface have been neglected, consistent with the theoretical results of Palmer (1976). Furthermore, variations in interfacial tension due to changes in surface temperature are ignored; that is, the Marangoni number is set equal to zero. Following Palmer (1976), the complete set of descriptive equations may be solved to yield a characteristic equation of the following form which defines the sufficient condition for interfacial instability in a rapidly evaporating liquid:

$$N_H = f_1 + f_2 N_p$$

where  $f_1$  and  $f_2$  are functions of  $\alpha$ ,  $N_{BO}$ ,  $N_{CR}$ ,  $N_{PR}$ ,  $N_{RE}$ ,  $N_p$ , and  $N_\mu$ . Stability criteria are established by locating the minimum in the curve of  $N_H$  vs. wave number  $\alpha$  for a given set of values for the other dimensionless groups.

## RESULTS

Figure 1 presents a representative graph of the critical Hickman number as a function of liquid phase Reynolds number for various values of  $N_p$  with  $N_p = 10^8$ . Notice that for  $N_p \geq 10^{-8}$ , an increase in  $N_p$  increases the stability limit for the system. Furthermore, as  $N_p$  is increased, the value of  $N_{RE}$  required for fluid-inertia instability (the point at which  $N_H$  drops sharply to zero)

\* For details of the mathematical analysis please see paragraph re Supplementary material on p. 186.

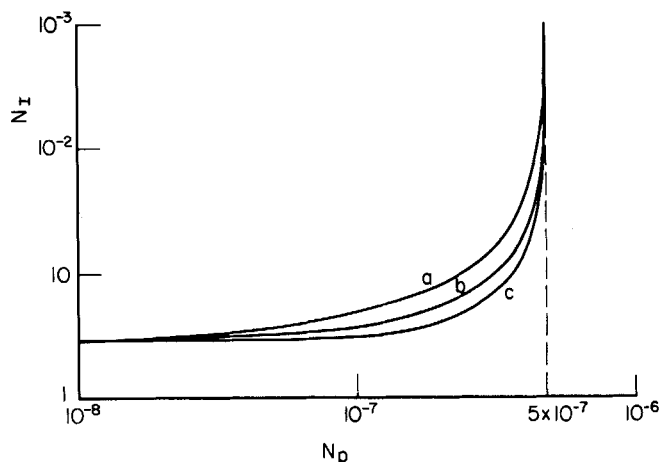


Fig. 2. The effect of pressure number on the criterion for fluid-inertia instability.  $N_{PR} = 10$ ,  $N_\mu = 10^2$ ,  $N_{BO} = 1$ ,  $N_{CR} = 10^{-5}$ . For curves (a)-(c),  $N_p = 10^8$ ,  $10^7$  &  $10^6$ , respectively.

gradually increases until at  $N_p \sim 5 \times 10^{-7}$  the potential for fluid-inertia instability is eliminated. Consequently, the range of  $N_{RE}$  for which vapor-recoil instabilities may exist is expanded substantially. While Figure 1 presents results for  $N_p = 10^8$ , the trends reflected in Figure 1 are seen for all values of  $N_p$ . In particular, whereas Palmer (1976) had shown that vapor-recoil instabilities are unlikely at pressures for which  $N_p = 10^8$ , the present results now indicate that instabilities via the vapor-recoil mechanism can be produced for  $N_{RE} > 0.08$  as long as  $N_p$  is greater than  $5 \times 10^{-7}$ .

The stability criterion for fluid-inertia instability is best characterized in terms of the inertial number  $N_I = N_{RE}^2 N_{PR} N_{CR} (N_p - 1)$  (see Palmer, 1976). Figure 2 presents the critical value of  $N_I$  vs.  $N_p$  for three values of  $N_p$ , with  $N_H = 0$ . Clearly, pressure effects on evaporation rate exert no significant influence on system stability for  $N_p < 10^{-8}$ . However, for  $N_p > 10^{-8}$ , the critical value of  $N_I$  increases rapidly until at  $N_p = 5 \times 10^{-7}$  the value of  $N_I$  approaches infinity. Furthermore, the stability limit is insensitive to the value of  $N_p$  and thus will not be influenced greatly by the operating pressure of an evaporation experiment. These results are particularly interesting because previously it had been believed that the upper bound on the stability of surfactant contaminated systems was defined by the criterion for fluid-inertia instability.

The value of  $N_p$  for a real system can vary widely depending on the physical properties of the liquid and the operating pressure for the evaporation experiment. In the case of triethanolamine (TEA) evaporating at a rate of  $5 \times 10^{-4}$  g/cm<sup>2</sup> s and with  $T \approx 120^\circ\text{C}$ ,  $N_p = 10^{-4}/P$  with  $P$  measured in millitorr. Therefore, for  $P < 200$  mtorr, interfacial instability via the fluid-inertia mechanism should not occur.

In conclusion, one qualification of this analysis needs to be mentioned. The analysis presumes that the gas phase is a continuum, an assumption which is not valid for all low pressure vaporization experiments. In particular, if the mean free path  $\lambda$  of the gas molecules is much greater than the length scale for the system  $\delta$ , then the concept of local pressure variations on the surface producing fluid-inertia instability and moderating local evaporation rates is physically unrealistic. For example, for TEA,  $\lambda \approx (4/P)$  cm. Therefore, the analysis certainly may be applied to TEA experiments in which  $\delta \geq 0.1$  cm and  $P > 400$  mtorr. Experiments are currently underway to determine low pressure limits on the applicability of the theoretical analysis.

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## NOTATION

$B$  = displacement of interface from its equilibrium position  
 $g$  = gravitational acceleration  
 $N_{BO}$  = Bond number =  $\delta^2 g (\rho_L - \rho_v) / \sigma$   
 $N_{CR}$  = Crispation number =  $\mu_{LK} / \sigma \delta$   
 $N_H$  = Hickman number =  $-(\partial \eta / \partial T) \eta \delta \Delta T \mu_v (\rho_v^{-1} - \rho_L^{-1}) / \rho_L \kappa_L \sigma$   
 $N_p$  = pressure number =  $-(\partial \eta / \partial P_v) \eta \mu_L \mu_v (\rho_v^{-1} - \rho_L^{-1}) / \rho_L \sigma \delta$   
 $N_{PR}$  = Prandtl number =  $\mu_L / \rho_L \kappa_L$   
 $N_{RE}$  = Reynolds number =  $\eta \delta / \mu_L$   
 $N_\rho$  = density ratio =  $\rho_L / \rho_v$   
 $N_\mu$  = viscosity ratio =  $\mu_L / \mu_v$   
 $P$  = pressure  
 $T$  = temperature

## Greek Letters

$\alpha$  = dimensionless wave number of the disturbance  
 $\delta$  = depth of the thermal boundary layer  
 $\eta$  = mass flux of evaporation

$\kappa$  = thermal diffusivity  
 $\mu$  = fluid viscosity  
 $\rho$  = fluid density  
 $\sigma$  = surface tension

## Subscripts

$L$  = liquid phase  
 $v$  = vapor phase

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Supplementary material has been deposited as Document No. 03285 with the National Auxiliary Publications Service (NAPS), c/o Microfilm Publications, 4 North Pearl Street, Portchester, N.Y. 10573, and may be obtained for \$3.00 for microfiche or \$5.00 for photocopies.

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# Stability of Liquid Films Method of Quadrature by Differentiation

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The flow of a thin liquid film down an inclined plane is frequently used as an idealized hydrodynamic description of the complex flow occurring in numerous industrial apparatuses (evaporators, condensers, rocket engines, nuclear reactors) and in some natural processes (soil erosion, transport of materials by rain water, spacecraft thermal shield melting). For a complete description, the stability of the interface has to be investigated because of the profound effect the waves may have on heat and mass transfer rates. The linear stability analysis of a thin liquid film flow by the perturbation method leads to an eigenvalue system. This system includes the well known Orr-Sommerfeld differential equation and four boundary conditions. Many attempts have been made to solve this system. The analytical studies of Yih (1955), Benjamin (1957), Yih (1963), Anshus and Goren (1966), Lin (1967), Krantz and Goren (1971), Krantz and Owens (1973), and Shuler and Krantz (1976) and the numerical calculations of Whitaker (1964), De Bruin (1974), and Sterling and Towell (1965) may be mentioned, in particular. However, we note that none of the analytical approaches offers a general solution; they are either limited in their field of application or based on approximations which are not always justified.

The purpose of the following is to find an analytical solution for the previous eigenvalue system which is valid

for not only small values of the wave number ( $\alpha < 0.3$ ) and which simply requires an analytic stream function on the interval  $[0;1]$ . This solution is based upon the method of quadrature by differentiation (Lanczos, 1956).

## FORMULATION OF THE PROBLEM

The temporal formulation of the well-known dimensionless Orr-Sommerfeld equation is given by

$$\phi^{iv} - 2\alpha^2 \phi'' + \alpha^4 \phi = i\alpha Re [(\bar{u} - c)(\phi'' - \alpha^2 \phi) - \bar{u}'' \phi] \quad (1)$$

The boundary conditions as formulated by Benjamin (1957) are

$$\phi'(1) = 0 \quad (2)$$

$$\phi(1) = 0 \quad (3)$$

$$\phi''(0) + \left(\alpha^2 - \frac{3}{c'}\right) \phi(0) = 0 \quad (4)$$

$$\frac{\alpha(3 \cot \theta + \alpha^2 We Re)}{c'} \phi(0)$$

$$+ \alpha(Re c' + 3\alpha i) \phi'(0) - i\phi'''(0) = 0 \quad (5)$$

with

$$c' \triangleq c - \frac{3}{2}; \quad \bar{u} = \frac{3}{2}(1 - y^2) \quad (6)$$