the soluble surfactant case is that Lin apparently expanded the surface concentration gradient in terms of α, whereas the work of Whitaker and Jones made a rather careful point of noting that the soluble surfactant case had to be treated in terms of an expansion in the square root of α. This becomes clear if one notes that the expression for the surface concentration gradient for a soluble surfactant contains the square root of α in the denominator.* Under these circumstances we are dealing with a function of the form,

$$\psi = \frac{A}{B + \sqrt{\alpha}} \tag{10}$$

and a Taylor series expansion in terms of α leads to

$$\psi(\alpha) = \psi|_{\alpha=0} + \alpha\left(\frac{\partial\psi}{\partial\alpha}\right)_{\alpha=0} + \frac{\alpha^2}{2}\left(\frac{\partial^2\psi}{\partial\alpha^2}\right)_{\alpha=0} + \ldots \tag{11}$$

One quickly sees that

$$\left(\frac{\partial\psi}{\partial\alpha}\right)_{\alpha=0} = \left(\frac{\partial^2\psi}{\partial\alpha^2}\right)_{\alpha=0} = \left(\frac{\partial^n\psi}{\partial\alpha^n}\right)_{\alpha=0} = \infty \tag{12}$$

thus ψ must be expanded in terms of $\sqrt{\alpha}$ not α. Lin apparently circumvented this difficulty by expanding the numerator of the term representing the surface concentration gradients, but not the denominator. Under these circumstances it is not surprising that the results of Lin are different from those of Whitaker and Jones.

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* See, for example, the equation for Σ' following Lin's Equation (13).

Comments on Surface Boundary Conditions for Small Amplitude Waves on a Falling Liquid Film

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The treatment of the boundary conditions for the problem of the stability of a contaminated liquid film by Lin (1970) is different from that by Whitaker and Jones (1966). Aside from the fundamental differences in the modeling of the monolayer, the following differences in the treatment of the boundary conditions are important. The problem of the present interest, in terms of mathematics, is a linear homogeneous eigenvalue problem. Hence, the eigenfunction can be obtained only up to an arbitrary constant multiplier. While the normalization of the eigenfunction is effected by Lin with $F(0) = 1$, the normalization by Whitaker and Jones is based on $F(0) = C - 1$. The latter normalization has one disadvantage in that it depends on the unknown eigenvalue C. Therefore, the normalization must be effected in each step of the perturbation solution. Consequently, the eigenvalue must be obtained in each step from five simultaneous algebraic equations instead of from four equations, which is the case in Lin's treatment. Moreover, the term $i\alpha^3 N_{Re} N_{We}$ in Equation (16) of Whitaker and Jones is treated as a term of $O(\alpha^3)$. Therefore, this term is not retained in their first three order perturbations. As is pointed out by Krantz and Goren (1970), this surface tension term is of order α according to the laboratory observations and must be included in the solution. As a direct consequence of this omission, C_1 given in (36) of Whitaker and Jones differs from that given in (16) of Lin by a term which represents the effect of surface tension. Indeed, the critical Reynolds number given in both papers are identical. This is due to the fact that the linear theory predicts the instability to occur as an infinitely long wave for which the surface tension cannot exhibit its effects and the erroneous omission of the surface tension term cannot be traced. The boundary conditions (16) and (17) with the additional condition (12) of Whitaker and Jones are equivalent to but not identical to the boundary conditions (8) and (9) of Lin. Thus, the discrepancy between the wave speeds obtained by Benjamin (1964) and Whitaker and Jones is not due to the incorrect boundary conditions. The difference in the real part of the wave speed is due to the fact that Benjamin did not carry the computation as far as Whitaker and Jones did. The difference in the imaginary part of the wave speed is due to the fact that Whitaker and Jones neglected the effect of surface tension in the first three order solutions.

For the case of soluble surfactants, Whitaker and Jones observe that $\alpha^{1/2}$ appears in the boundary condition. This leads them to expand the eigenfunction and the eigenvalue in power series of $\alpha^{1/2}$. The solution is obtained up to $\alpha^{5/2}$-term. There exists no mathematical theory which ensures the convergence of the perturbation solution either in powers of $\alpha^{1/2}$ or α. The perturbation solution can best be regarded as an asymptotic solution. Note that there is a singularity in the expression of Σ' at $\alpha = 0$. Therefore a single expansion which is uniformly valid for $0 \leqq \alpha << 1$ is not possible. It was assumed that $s_0/\alpha^{1/2} = O(1)$ in the boundary condition (13) of Lin's work. Thus, the expansion of Σ' mentioned by Whitaker is not necessary. Obviously, the results obtained on the basis of this assumption do not apply to the physically meaningless case of $\alpha = 0$ and to the case of strong solubility $s_0 >> O(\alpha^{1/2})$. However, there was an algebraic error in (20) and (21) of Lin. With this correction, these equations read

$$\Delta c_i = i\alpha \left[\Sigma N_{Re} \frac{1 - (N_{Pe}\alpha/3)^{1/2}/E}{\{1 + (N_{Pe}\alpha/3)^{1/2}/E\}^2 + N_{Pe}\alpha/3E^2} + \frac{6}{5} N_{Re} - \frac{1}{3}\,(3\cot\beta + \alpha^2 S N_{Re}) \right],$$

$$\Delta c_r = \alpha \Sigma N_{Re}\,(\alpha N_{Re}/3)^{1/3}/E \ .$$

Nevertheless, the conclusion is not affected. Whitaker and Jones' expansion has some disadvantages as are pointed out by they themselves: "Unfortunately, this provides us with a slowly converging perturbation solution, and in addition there is a singularity in the expansion of $\partial\Gamma/\partial X$ for $\kappa \rightarrow \infty$." Lin obtains the solution only up to $O(\alpha)$. However, higher order solutions are of little practical significance since the effect of the wave amplitude (which is neglected in the linear theory) is at least of the comparable order of magnitude as the higher order term as can be seen from the work of Krantz and Goren (1970), Javdani and Goren (1971), and Lin (1969, 1970a, 1970b, 1971).

In conclusion, the limitation of the linear stability theory is pointed out. It is assumed in the linear theory that the wave speed and the wave length are independent of the wave amplitude, and that Fourier components of the disturbance do not interact. This is clearly an unrealistic assumption especially when it is applied to the film flow. As a consequence of this unrealistic assumption, the linear theory predicts the instability to occur as an infinitely long gravity wave. On the other hand, only finite amplitude waves with finite lengths are actually observed in laboratories. Although this has led many workers to speculate that the observed waves correspond to the most amplified wave, the fact remains that the observed waves are of finite amplitude which do not grow exponentially. All evidence clearly indicate that wavy film flows are inherently nonlinear. An intelligent design of any industrial apparatus involving wavy film flows must be based on a sound understanding of the nonlinear mechanism. For this purpose, the nonlinear analysis must include at least the effects of the amplitude and the modal interactions. A nonlinear analysis which includes the effect of modal interactions in a viscous film with surface tension is not yet available.

NOTATION

c = complex growth rate constant, cm/s
C = c/u_0, dimensionless complex growth rate constant
f = stream function, cm^2/s
F = f/u_0h, dimensionless stream function
h = film thickness, cm
K = equilibrium coefficient, cm
u_0 = surface velocity, cm/s
x = spatial coordinate, cm
X = x/h, dimensionless spatial coordinate

Greek Letters

α = $2\pi h/\lambda$, dimensionless wave number
Γ = dimensionless surface density = Σ'
ν = kinematic viscosity, cm^2/s
σ = surface tension, dyne/cm
ρ = fluid density, g/cc

Dimensionless Groups

κ = K/h, dimensionless equilibrium constant
N_{Re} = u_0h/ν, Reynolds number
N_{We} = $\sigma/\rho u_0^2h$, Weber number

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