To illustrate the proposed method, Figure 1 shows calculated and experimental liquid thermal conductivity data for the system methanol water at 0° and 60°C. Figure 2 exemplifies a limitation of Equation (1) in that it fails to predict the minimum thermal conductivities for the azeotropic liquid mixture, CCl_4/tert-butanol.

DISCUSSION

The size and the number of molecules are equally important. This can be verified by neglecting the size effect when the mole fraction is used instead of the volumetric fraction in Equation (1). If such a substitution is made, the error in the calculated values will be significantly greater than for calculations with the volumetric average used. This indicates that for energy transport in a liquid mixture, the volumetric fraction is more appropriate than the molar fraction. Equation (1), however, reduces to a molar average when the liquid molar volumes of the components are equal.

The proposed method fails to predict any minimum or maximum thermal conductivity of a mixture as a function of composition. This limitation is shown in Figure 2 for the azeotropic liquid mixture carbon tetrachloride-*t*-butanol.

To compare the accuracy of the new method with others that have been proposed, in Table 4, we show calculated absolute percent deviations from experimental data. The column marked harmonic indicates that Equations (1) through (4) of the present Note were used. Also shown are results assuming a geometric mean and an arithmetic for λ_{ij}; that is

$$\lambda_{ij} = \sqrt{\lambda_i \lambda_j} \qquad \text{geometric mean} \tag{5}$$

$$\lambda_{ij} = (\lambda_i + \lambda_j)/2 \qquad \text{arithmetic mean} \tag{6}$$

Clearly, the harmonic mean choice leads to the least error.

Also shown are deviations for the NEL correlation (Jamieson and Hastings, 1969) for binary systems

$$\frac{\lambda_m - \lambda_1}{\lambda_2 - \lambda_1} = \alpha w_2^{3/2} + w_2(1 - \alpha) \tag{7}$$

where w_2 is the weight fraction of component 2 and $\lambda_2 > \lambda_1$. The adjustable parameter α was set equal to unity. In addition, the Filippov relation was tested (Filippov, 1955):

$$\frac{\lambda_m - \lambda_1}{\lambda_2 - \lambda_1} = C w_2^2 + w_2(1 - C) \tag{8}$$

w_2 was defined above, and the parameter C was chosen to equal 0.7 (Reid et al., 1976). As suggested by Reid (1976), if Equation (1) is used with the geometric mean assumption for λ_{ij} [Equation (5)] and compared to Equation (8), it can be shown that

$$C = \left| \frac{\lambda_2^{1/2} - \lambda_1^{1/2}}{\lambda_2^{1/2} + \lambda_1^{1/2}} \right| \tag{9}$$

The conclusion from the results shown in Table 4 is that the harmonic mean assumption [Equations (1) through (4)] yields estimations of λ_m which are generally more accurate than other correlations and within normal experimental error (circa ±3%).

ACKNOWLEDGMENT

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NOTATION

V = liquid molar volume
w = weight fraction
x = mole fraction
ϕ = volume fraction
λ = thermal conductivity

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The Linear Hydrodynamic Stability of Film Flow Down a Vertical Cylinder

WILLIAM B. KRANTZ and RICHARD L. ZOLLARS

Department of Chemical Engineering
University of Colorado
Boulder, Colorado 80302

Film flow occurs in a variety of engineering equipment for gas-liquid contacting and in a number of industrial processes for coating and surface treating solid surfaces. Furthermore, wetted wall columns are widely used as laboratory absorbers and reactors for determining mass transfer coefficients and kinetic data. Engineers have been interested in the stability of this flow because of its pro-

Correspondence concerning this paper should be addressed to W. B. Krantz. R. L. Zollars is with Union Carbide Corporation, South Charleston, West Virginia 25303.

pensity to develop ripples even at very small Reynolds numbers. In many industrial flows, these ripples are beneficial since they increase the transfer coefficients. However, in other cases, such as coating by extrusion and withdrawal processes, they are detrimental and may result in product nonuniformity. In particular, the occurrence of ripples is disadvantageous in laboratory absorbers and reactors since it results in overestimating the mass transfer coefficients.

Most solutions to this stability problem have been confined to planar film flows, although many industrial applications as well as laboratory absorbers involve flow over surfaces with small radii of lateral curvature. Surprisingly, these predictions for the stability of planar film flow frequently have been tested by using cylindrical wetted wall columns without assessing the significance of the lateral curvature effects. The latter effects are considered in this note which develops an asymptotic solution to the linear stability problem for film flow down a vertical cylinder.

RELATED THEORETICAL STUDIES

Hasegawa and Nakaya (1970) analyzed this problem via an approximate numerical solution. Zollars (1974) has indicated that there appear to be several nontrivial errors in their analysis.

Hickox (1971) considered the stability of two viscous immiscible fluids flowing concentrically in a vertical cylindrical tube. However, his analysis precludes considering a viscous liquid in contact with an inviscid gas which is of interest here.

Lin and Liu (1975) developed an asymptotic solution for the linear stability of film coating of wires and tubes. Their analysis, while substantially correct and of considerable value, does not place proper bounds on the domain of validity of their solution. Consequently, some of their results shown graphically are for conditions which violate the proper ordering arguments for their solution. Lin and Liu also consider the implications of their solution for the rather interesting case of no-flow. However, their results do not agree with those of Goren (1962) who employed a systematic analytical solution to show that a nonflowing liquid coating on a cylinder was unstable owing to the lateral curvature effects. In addition, their results imply that unstable disturbances on a nonflowing liquid film have a nonzero phase velocity. A partial explanation for these anomalous predictions is that the no-flow case is an improper limit for the scaling employed by Lin and Liu. The perturbation velocities and complex wave velocity are nondimensionalized with the surface velocity and thus are not well behaved in the limit of no-flow. Whereas the surface velocity is zero for the no-flow case, the perturbation velocities and wave velocity are nonzero.

In this note an asymptotic solution will be developed which closely parallels that of Lin and Liu. However, this solution differs from that of Lin and Liu in that the stability problem is scaled appropriate to conditions such that the capillary pressure forces balance the viscous forces. This results in dimensionless variables which are well behaved in the limit of no-flow. In addition, the domain of validity of the resulting asymptotic solution is established. Lastly, the error incurred in those experiments which employed cylindrical wetted wall columns to test linear stability theory for planar film flows is assessed.

THEORETICAL DEVELOPMENT

Consider viscous film flow down a vertical cylidner of radius R. Let x be the streamwise coordinate and y the cross-stream coordinate such that $y = 0$ defines the surface of the cylinder. The appropriate form of the temporal growth formulation of the dimensionless Orr-Sommerfeld equation is given by

$$\begin{aligned}\phi'''' &- \frac{2}{(\Lambda+y)}\phi''' - 2\alpha^2\phi'' + \frac{3}{(\Lambda+y)^2}\phi'' \\ &+ i\alpha N_{Oh}^{-2}c\phi'' - \frac{3}{(\Lambda+y)^3}\phi' + \frac{2}{(\Lambda+y)}\alpha^2\phi' \\ &- \frac{1}{(\Lambda+y)}i\alpha N_{Oh}^{-2}c\phi' + \alpha^4\phi - i\alpha^3 N_{Oh}^{-2}c\phi \\ &= N_{Re}\bar{u}i\alpha\phi'' - \frac{N_{Re}\bar{u}}{(\Lambda+y)}i\alpha\phi' - N_{Re}i\alpha\bar{u}''\phi \\ &+ \frac{N_{Re}\bar{u}'}{(\Lambda+y)}i\alpha\phi - N_{Re}\bar{u}i\alpha^3\phi \end{aligned} \quad (1)$$

This is subject to the boundary conditions

$$\phi' = 0 \quad @ \quad y = 0 \quad (2)$$

$$\phi = 0 \quad @ \quad y = 0 \quad (3)$$

$$\frac{N_{Re}\bar{u}''\phi}{(N_{Re}\bar{u} - N_{Oh}^{-2}c)} - \phi'' + \frac{1}{(\Lambda+1)}\phi' - \alpha^2\phi = 0 \quad @ \quad y = 1 \quad (4)$$

$$\begin{aligned}\phi''' &- \frac{1}{(\Lambda+1)}\phi'' + \frac{1}{(\Lambda+1)^2}\phi' - 3\alpha^2\phi' \\ &+ i\alpha N_{Oh}^{-2}c\phi' - i\alpha N_{Re}\bar{u}\phi' \\ &+ \frac{2\alpha^2}{(\Lambda+1)}\phi + \frac{i\alpha^3 N_{Oh}^{-2}}{(N_{Re}\bar{u} - N_{Oh}^{-2}c)}\phi \\ &- \frac{i\alpha N_{Oh}^{-2}}{(\Lambda+1)^2(N_{Re}\bar{u} - N_{Oh}^{-2}c)}\phi = 0 \end{aligned} \quad @ \quad y = 1 \quad (5)$$

where the basic flow velocity $\bar{u}$ is given by

$$\bar{u} = \eta\left[2(\Lambda+1)^2 \ln\left(\frac{\Lambda+y}{\Lambda}\right) - 2\Lambda y - y^2\right] \quad (6)$$

and

$$\frac{1}{\eta} = \left[2(\Lambda+1)^2 \ln\left(\frac{\Lambda+1}{\Lambda}\right) - 2\Lambda - 1\right] \quad (7)$$

In arriving at this dimensionless formulation, the linearized equations of motion and appropriate boundary conditions have been scaled such that the pressure, surface tension, and viscous forces are of the same order. This scaling implies that the perturbation velocities and complex wave velocity c are nondimensionalized with respect to σ/μ, x, and y with respect to $\bar{h}$, and t with respect to h^2/ν; note, however, that the basic flow velocity is nondimensionalized with respect to $\bar{u}_s$, the surface velocity of the basic flow. This scaling introduces the Reynolds number $N_{Re} = \bar{u}_s\bar{h}/\nu$; Ohnesorge number $N_{Oh} = \mu/(\rho\sigma h)^{1/2}$, a measure of the viscous to surface tension forces; and curvature group $\Lambda = R/\bar{h}$.

We seek an asymptotic solution to the above for long waves; hence

$$\phi = \phi_0 + \phi_1\alpha + o(\alpha^2) \quad (8)$$

The zeroth-order system of equations to be solved is then given by

$$\phi_0'''' - \frac{2}{(\Lambda+y)}\phi_0''' + \frac{3}{(\Lambda+y)^2}\phi_0''$$

$$-\frac{3}{(\Lambda+y)^3}\phi_0' = 0 \quad (9)$$

$$\phi_0' = 0 \quad @ \quad y = 0 \quad (10)$$

$$\phi_0 = 0 \quad @ \quad y = 0 \quad (11)$$

$$\phi_0'' - \frac{1}{(\Lambda+1)}\phi_0' + \frac{4\eta N_{Re}}{(N_{Re} - N_{Oh}^{-2}c_0)}\phi_0 = 0 \quad @ \quad y = 1 \quad (12)$$

$$\phi_0''' - \frac{1}{(\Lambda+1)}\phi_0'' + \frac{1}{(\Lambda+1)}\phi_0' = 0 \quad @ \quad y = 1 \quad (13)$$

The first-order system of equations is given by

$$\phi_1'''' - \frac{2}{(\Lambda+y)}\phi_1''' + \frac{3}{(\Lambda+y)^2}\phi_1'' - \frac{3}{(\Lambda+y)^3}\phi_1'$$
$$= i\left\{(N_{Re}\bar{u} - N_{Oh}^{-2}c_0)\left[\phi_0'' - \frac{1}{(\Lambda+y)}\phi_0'\right] - N_{Re}\left[\bar{u}'' - \frac{\bar{u}'}{(\Lambda+y)}\right]\phi_0\right\} \quad (14)$$

$$\phi_1' = 0 \quad @ \quad y = 0 \quad (15)$$

$$\phi_1 = 0 \quad @ \quad y = 0 \quad (16)$$

$$\phi_1'' - \frac{\phi_1'}{(\Lambda+1)} + \frac{4\eta N_{Re}N_{Oh}^{-2}\phi_0 c_1}{(N_{Re} - N_{Oh}^{-2}c_0)^2} + \frac{4\eta N_{Re}}{(N_{Re} - N_{Oh}^{-2}c_0)}\phi_1 = 0 \quad @ \quad y = 1 \quad (17)$$

$$\phi_1''' - \frac{\phi_1''}{(\Lambda+1)} + \frac{\phi_1'}{(\Lambda+1)^2} - i(N_{Re} - N_{Oh}^{-2}c_0)\phi_0' + \frac{iN_{Oh}^{-2}}{(N_{Re} - N_{Oh}^{-2}c_0)}\left[\alpha^2 - \frac{1}{(\Lambda+1)^2}\right]\phi_0 = 0 \quad @ \quad y = 1 \quad (18)$$

In arriving at Equations (9) through (18) we have made the following ordering arguments:

$$1/\Lambda = O(1) \quad (19)$$

$$N_{Re} = O(1) \quad (20)$$

$$N_{Oh} = O(\alpha) \quad (21)$$

$$N_{Oh} = O[1/(\Lambda+1)] \quad (22)$$

Note that the symbol O, for example in $N_{Re} = O(1)$, implies that the Reynolds number is at most of order unity. The provocation for these ordering arguments should be obvious. For example, if $1/\Lambda = O(1)$ did not apply, then the terms proportional to $(\Lambda + y)^{-1}$ in Equation (1) could become the dominant terms. The other ordering arguments stem from similar considerations.

Equation (9) and the homogeneous part of Equation (14) are Cauchy equations and are readily solvable. The resulting eigenvalue problem yields an equation for the complex eigenvalue $c = c_r + ic_i$ in terms of the parameters of the problem:

$$c_r = 2N_{Re}N_{Oh}^2 \quad (23)$$

$$\alpha c_i = (f_1\eta^2/8)N_{Re}N_{Oh}^2\alpha + f_2\alpha[\alpha^2 - 1/(\Lambda+1)^2] \quad (24)$$

where

$$f_1 = 16(\Lambda+1)^6\left[\ln\left(\frac{\Lambda+1}{\Lambda}\right)\right]^3 - (40\Lambda^5 + 180\Lambda^4 + 320\Lambda^3 + 280\Lambda^2 + 120\Lambda + 20\left[\ln\left(\frac{\Lambda+1}{\Lambda}\right)\right]^2 - (20\Lambda^6 + 180\Lambda^5 + 190\Lambda^4 + 136\Lambda^3 + 34\Lambda^2)\ln\left(\frac{\Lambda+1}{\Lambda}\right) + 20\Lambda^5 + 98\Lambda^4 + \frac{500}{3}\Lambda^3 + 140\Lambda^2 + 59\Lambda + \frac{59}{6} \quad (25)$$

$$f_2 = \frac{(2\Lambda+1)^2}{16(\Lambda+1)} - \frac{\Lambda+1}{8n} \quad (26)$$

If Equations (23) and (24) are recast in terms of Lin and Liu's dimensionless variables, they agree identically. In the limit of $\Lambda \to \infty$, Yih's (1963) results for planar film flow are recovered.

DISCUSSION

Note that Equation (24) governs the stability of this flow. If $\alpha c_i > 0$, an infinitesimal disturbance of wave number α will grow exponentially in time; if $\alpha c_i < 0$, this disturbance will decay; if $\alpha c_i = 0$, the disturbance is neutrally stable. The first and second terms in brackets in Equation (24) are the streamwise and lateral curvature effects, respectively. Our ordering argument on Λ implies that $f_1 > 0$, and $f_2 < 0$. Hence, the streamwise curvature of the perturbed flow is a stabilizing term. The lateral curvature term, on the other hand, is a destabilizing term. This destabilizing effect occurs because the troughs of the waves have a smaller radius of curvature than do the crests. This induces a capillary pressure force tending to move fluid from the troughs into the crests, thus promoting growth of the waves.

Neutral stability curves for a typical mineral oil (μ = 1.46 poise; ρ = 0.868 g/cm^3, α = 30.8 dynes/cm, at 25°C) and water are shown in Figures 1 and 2, respectively. These fluids are characterized by the surface tension group $N_\zeta = (\sigma/\rho)(2/g\nu^4)^{1/3} = (2\eta^{1/3}N_{Re}^{1/3}N_{Oh}^2)^{-1}$. Note that neutral stability curves for curvature groups less than unity and Reynolds numbers greater than unity are not shown in Figures 1 and 2 because of our ordering arguments given by Equations (19) and (20). All wave numbers below the neutral stability curves are unstable. Note that whereas planar film flow is unstable at all nonzero Reynolds numbers and neutrally stable at $N_{Re} = 0$, film flow down a cylinder is unstable at all Reynolds numbers. A decrease in the curvature group or surface tension group and an increase in Reynolds number are seen to be destabilizing.

It is of interest to assess the error incurred in those experimental studies which employed a cylindrical wetted wall column to test the predictions of linear stability theory for planar film flow. The smallest columns used in these studies had a radius of approximately 2.5 cm. The Reynolds numbers studied were in the range $6.6 \leq N_{Re} \leq 216$, with water used as the test fluid; indeed, it is experimentally impractical to operate at smaller Reynolds numbers with water because of the extremely thin films involved. Figure 2 for water indicates that for $N_{Re} = 0.1$ and Λ = 100, the neutrally stable wave number for the cylinder is 230% greater than that for the planar case. This corresponds to $\bar{h}$ = 0.00273 cm and R = 0.273 cm. At N_{Re} = 0.5 and Λ = 100, the neutrally stable wave number for cylinder is 36% greater. This corresponds to $\bar{h}$ = 0.00468 cm and R = 0.468 cm. Thus we can reasonably conclude that these experimental studies incurred little error in ignoring the lateral curvature effect. However, studies on

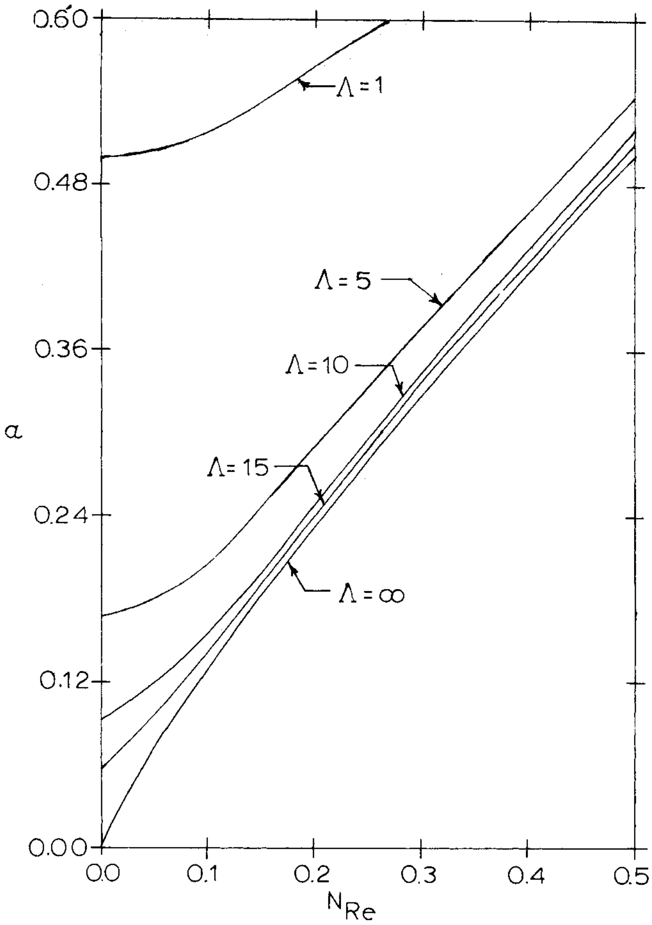

Fig. 1. Neutral stability curves for film flow down a vertical cylinder with $N_\zeta = 2.0$.

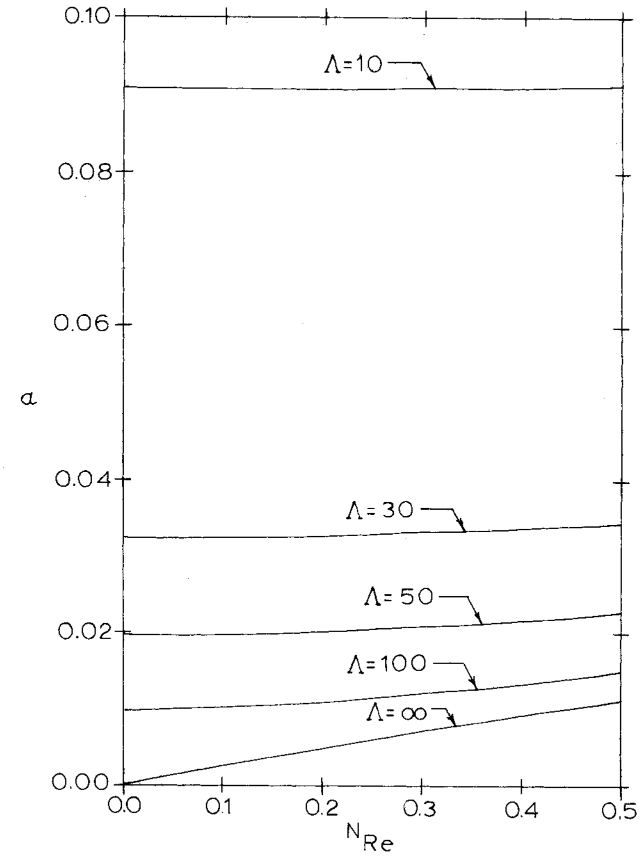

Fig. 2. Neutral stability curves for film flow down a vertical cylinder with $N_\zeta = 4\,200$.

cylindrical columns having radii as large as 2.5 cm could incur significant error by ignoring the lateral curvature effect at small Reynolds numbers if fluids were used having viscosities of the same order as that of the light mineral oil of Figure 1.

Figures 1 and 2 indicate that finite values of the curvature group lead to an unstable band of disturbances even at $N_{Re} = 0$. This corresponds to a nonflowing basic film supported by a cylindrical core. The dimensionless variables used here are well behaved in the limit of no-flow; thus, in the limit of zero Reynolds number, Equations (23) and (24) yield

$$c_r = 0 \tag{27}$$

$$\alpha c_i = f_2 \alpha[\alpha^2 - 1/(\Lambda + 1)^2] \tag{28}$$

Note that Lin and Liu predict that their dimensionless phase velocity is equal to two for all long waves, even at zero Reynolds number. The latter is impossible, since for the nonflow case the waves are stationary.

The results obtained here for zero Reynolds number can be compared with those of Goren (1962) who obtained a closed-form analytical solution for the stream function for this case. Equation (28) predicts a neutrally stable wave number $\alpha_n = 1/(\Lambda + 1)$ which agrees identically with that predicted by Goren. Although Goren obtained a closed-form analytical solution for the stream function, he could not solve the resulting eigenvalue problem for the amplification factor for nonneutrally stable waves except in the limiting cases of negligible inertia and negligible viscosity. He presents his results for these two limiting cases as a plot of the most highly amplified wave number vs. $R/(R + \overline{h}) = \Lambda/(\Lambda + 1)$. Since this problem is scaled such that the viscous forces balance the capillary pressure forces, our results can be compared only with Goren's negligible inertia results. Furthermore, the ordering argument on the curvature group given by Equation (19) implies that our results, as well as those of Lin and Liu, can be compared with Goren's only over the range $\frac{1}{2} \leq R/(R + \overline{h}) < 1$. We cannot compare results at $R/(R + \overline{h}) = 1$, since Goren achieves this limit by letting $\overline{h} \to 0$, whereas we achieve it by letting $R \to \infty$; our dimensionless variables are not well behaved in the limit of $\overline{h} \to 0$. Equation (28) then indicates that the most highly amplified wave number is given by $\alpha_m = 1/3^{1/2}(\Lambda + 1)$. Goren's solution for negligible inertia indicates that at $R/(R + \overline{h}) = \frac{1}{2}$, $\alpha_m = 0.447$, whereas our result yields $\alpha_m = 0.386$. The disparity between these two results is due to the fact that the solution developed here, as well as that of Lin and Liu, is restricted to $\alpha << 1$, whereas Goren's most highly amplified wave numbers for the case of negligible inertia are reasonably large.

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NOTATION

- c = complex wave velocity nondimensionalized with respect to σ/μ
- c_i = imaginary part of complex wave velocity
- c_k = k^{th}-order solution for complex wave velocity
- c_r = real part of complex wave velocity

f_1 = function defined by Equation (25)
f_2 = function defined by Equation (26)
g = gravitational acceleration
$\bar{h}$ = basic film thickness
i = $\sqrt{-1}$
N_{Oh} = Ohnesorge number = $\mu/(\rho\sigma\bar{h})^{1/2}$
N_{Re} = Reynolds number = $\bar{u}_s\bar{h}/\nu$
N_ζ = surface tension group = $(\sigma/\rho)(2/g\nu^4)^{1/3}$
R = radius of cylinder
t = time nondimensionalized with respect to $\bar{h}^2/\nu$
$\bar{u}$ = basic flow velocity nondimensionalized with respect to $\bar{u}_s$
$\bar{u}_s$ = surface velocity of basic flow
x = streamwise coordinate nondimensionalized with respect to $\bar{h}$
y = cross-stream coordinate nondimensionalized with respect to $\bar{h}$

Greek Letters

α = wave number = $2\pi\bar{h}/\lambda$
α_m = most highly amplified wave number
α_n = neutrally stable wave number
η = function defined by Equation (7)
λ = wavelength
Λ = curvature group = $R/\bar{h}$
μ = shear viscosity
ν = kinematic viscosity
ρ = density
σ = surface tension
ϕ = amplitude of stream function nondimensionalized with respect to $\sigma\bar{h}^2/\nu$
ϕ_k = k^{th}-order solution for amplitude of stream function

Superscripts

= order of differentiation with respect to y

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The Equivalence of the Spatial and Temporal Formulations for the Linear Stability of Falling Film Flow

PATRICK J. SHULER and WILLIAM B. KRANTZ

Department of Chemical Engineering
University of Colorado, Boulder, Colorado 80302

The tendency of falling film flow to be unstable at very low Reynolds numbers leads to the inevitable appearance of ripples. Engineers have been interested in the stability of this flow because of the profound effect these ripples can have on heat and mass transfer rates. The initial linear stability analyses of this flow were developed for temporally growing disturbances whose stream function is given by $\psi = \phi(y)\cdot\exp[i(\alpha_r x - \omega t)]$ and $\omega = \omega_r + i\omega_i$. Recently, this problem has been solved for spatially growing disturbances whose stream function is given by $\psi = \phi(y)\cdot\exp[i(\alpha x - \omega_r t)]$, where $\alpha = \alpha_r + i\alpha_i$.

The question arises as to whether the predictions of the spatial and temporal formulations are equivalent for all observable disturbances. Prior attempts to resolve this question have been inconclusive, as has been discussed by Lin (1975*a*) and Krantz (1975*a*, 1975*b*), either because they employ nonsystematic methods of solution whose range of validity cannot be assessed, or because they are restricted to weakly amplified disturbances for which a transformation of Gaster (1965) assures the equivalence of the two formulations. This transformation permits one to convert the temporal amplification factor into a corresponding spatial amplification factor. Gaster's development is in essence an expansion of the characteristic equation for the linear stability problem about the neutral stability condition, at which of course the predictions of the temporal and spatial formulations are identically equivalent. Thus, this transformation of Gaster is restricted to weakly amplified waves.

The purpose of this note is to compare the predictions of the spatial and temporal formulations of this linear stability problem solved via a systematic method of solution which is not necessarily restricted to weakly amplified long waves.

THEORETICAL DEVELOPMENT

The spatial formulation of the dimensionless Orr-Sommerfeld equation and associated boundary and kinematic conditions appropriate to falling film flow is given by

$$\phi'''' - 2\alpha^2\phi'' + \alpha^4\phi = i\alpha N_{Re}\left[\left(U - \frac{\omega_r}{\alpha}\right)(\phi'' - \alpha^2\phi) - U''\phi\right] \quad (1)$$

$$\phi = 0, \quad \text{at} \quad y = 1 \quad (2)$$

$$\phi' = 0, \quad \text{at} \quad y = 1 \quad (3)$$

$$\phi'' + \left[\alpha^2 - \frac{3}{\left(\dfrac{\omega_r}{\alpha} - \dfrac{3}{2}\right)}\right]\phi = 0, \quad \text{at} \quad y = 0 \quad (4)$$