# Various Types of Instabilities on a Falling Liquid Film

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The gravity driven flow of a thin liquid film is frequently encountered in chemical processing equipment. An early analysis by Nusselt of condensation heat transfer contains the essential laminar steady fluid mechanics pertaining to gravity-driven constant thickness film flow. In reality the flow of thin films is more complex because wave motion, more intense with increasing Reynolds numbers, occurs, and because dry patches may be generated at very low Reynolds numbers. The wave motion and the occurrence of dry patches are consequences of two different types of instability. The wave motion instability is a time-dependent surface-oriented instability which results, ultimately in traveling waves which are more intense at higher fluid velocities (Tailby and Portalski, 1961). On the other hand, experimental evidence obtained by Norman and McIntyre (1960) and by Simon and Hsu (1970) indicates that for nonwetting liquids, the film cannot be maintained continuous if the flow rate or average film thickness is less than some nonzero critical value. The critical film thickness, so defined, reported by Simon and Hsu (1970) for room temperature water flowing down an "unheated conducting glass tube" is $1.4 \times 10^{-4}$ m. This critical point increases with heat flux from the solid wall. The appearance of dry patches on the wall is preceded by a roping instability—a disturbance quasi-periodic in the cross-stream direction and growing or decaying in the downstream directions—visible on the film surface. Yet another instability observed by Gerstmann and Griffith (1967) is that which leads to longitudinal ridges or ribs on a viscous film flowing or draining down the underside of a solid surface. This instability, in contrast to the two discussed above, has a readily apparent physical explanation: Since the fluid is on the underside of the surface, it is gravitationally unstable.

In what follows, these three types of instability are subjected to a common treatment which yields and contrasts the essential features of each.

## GOVERNING EQUATIONS

### Mean Flow

A viscous, Newtonian fluid is assumed to flow, under the action of gravity, down a plane inclined at an angle $\beta$ to the horizontal. The steady Nusselt solution is found upon assuming that one surface is fixed and the other is free and that the film is of constant thickness. Then the velocity $\overline{U}$ in the $x$-direction and the isotropic pressure $P$ are given by

$$\overline{U} = \frac{3}{2}\overline{U}_a\left(1 - \frac{Y^2}{h^2}\right) \qquad (1)$$

$$P = P_0 + \rho g Y \cos\beta \qquad (2)$$

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### Linear Stability Equations

In a linear stability analysis the mean flow is perturbed infinitesimally and the subsequent growth or decay is determined from the eigenvalues of the linear homogeneous equations which result. If any disturbance on a flow results in an eigenvalue which predicts growth, the flow is said to be unstable to that disturbance. If all disturbances decay, the flow is said to be linearly stable. Anticipating that the disturbance equations are linear and, because of the mean flow, have coefficients which only depend on the depth $Y$ the form of the disturbance variables ($U_x'$, $U_y'$, $U_z'$, $P'$, $\eta'$) in nondimensional form is assumed to be

$$f'(x, y, z, t) = \hat{f}(y)\exp(inz + mx + \gamma t) \qquad (3)$$

This represents but one term of a Fourier expansion of a disturbance variable. The disturbance form as written is intended to allow for periodicity in the $z$ or cross-stream direction (that is, $n$ is real), growth and/or periodicity in the $x$ or stream-wise direction ($m$ is complex), and growth and/or periodicity in time ($\gamma$ is complex). The resulting homogeneous ordinary differential equation is then

$$(D^2 + m^2 - n^2)^2\hat{v} = mRe\left[\left(\overline{u} + \frac{\gamma}{m}\right)(D^2 + m^2 - n^2)\hat{v} - \hat{v}D^2\overline{u}\right] \qquad (4)$$

Equation (4) is to be solved subject to four boundary conditions which result from the condition of zero velocity at the wall, a tangential stress balance at the free surface, and the balance of normal fluid stresses including the surface tension-curvature effect at the free surface. In addition, there is a kinematic condition which equates the time rate of change of surface position $d\eta'/dt$ to the fluid velocity $U_y'$ at the surface. The resulting boundary conditions are, in the order named above,

$$\hat{v} = D\hat{v} = 0 \quad \text{at} \quad y = 1 \qquad (5)$$

$$(D^2 - m^2 + n^2)\hat{v} - \frac{D^2\overline{u}}{u + \dfrac{\gamma}{m}}\hat{v} = 0 \quad \text{at} \quad y = 0 \qquad (6)$$

$$\left[D^2 + 3(m^2 - n^2) - mRe\left(\overline{u} + \frac{\gamma}{m}\right)\right]D\hat{v} + \frac{m^2 - n^2}{m\left(\overline{u} + \dfrac{\gamma}{m}\right)}\left[-3\cot\beta + (m^2 - n^2)\zeta Re^{-2/3}\right]\hat{v} = 0 \quad \text{at} \quad y = 0 \qquad (7)$$

Depending upon the nature of the perturbation we can have the following situations: (1) A traveling wave in the stream-wise direction with an exponential time dependent amplitude (in this case $n = 0$, $m_{real} = 0$); this kind of perturbation leads to wave motion. (2) A time steady perturbation growing or decaying in the downstream direction involving the thinning of the film in some regions and a corresponding thickening in neighboring regions (in this case $m_{imag} = 0$, $\gamma = 0$); this kind of perturbation leads, for $0 < \beta \leq 90°$, to dry patches and, for $90° < \beta < 180°$, can predict ribs on the underside of a surface. Several other situations are possible. In addition, let us mention the perturbation in the form of a traveling wave in the streamwise direction with the amplitude spatially growing or decaying. This leads also to wave motion as shown by Krantz and Owens (1973).

*Case i* $m_{real} = 0$, $n = 0$—*traveling waves with time-dependent amplitude,* To make this section consistent with other works on the subject, the parameters are renamed $m_{imag} \equiv \alpha$ and $\gamma \equiv -i\alpha c$. One recovers from Equations (4) to (7) the set of equations found for the study of waves on a falling film (Benjamin, 1957; Yih, 1963; or Anshus, 1972). The criterion for instability is that $c_{imag}$ is greater than zero, and $c_{real}$ is then the (nondimensional) velocity of the traveling wave. Because the solution to the corresponding equations is developed in such detail elsewhere (Yih, 1963), it won't be developed here. For the present purpose the essential quantitative result, reported by Yih (1963), is that to lowest order in small wave number $\alpha$ the wave velocity is three times the average film velocity and the growth eigenvalue is given by

$$\gamma_{real} = \alpha c_{imag} = \alpha^2 \left( \frac{6Re}{5} - \cot\beta - \frac{\alpha^2}{3} \zeta Re^{-2/3} \right) \quad (8)$$

Thus the critical Reynolds number, above which the flow is unstable to a traveling wave disturbance is $Re_c = 5/6 \cot\beta$, and above $Re_c$ the growth parameter depends on $\alpha$ and decreases as $\zeta$ increases or $Re$ decreases. If $\cot\beta = 0$ the flow is unstable even down to zero Reynolds number.

*Case ii* $m_{imag} = 0$, $\gamma = 0$—*dry patches, time independent perturbation.* Again the parameters are renamed, $m_{real} \equiv a$ and $n \equiv \alpha$. One recovers the set of real equations examined recently by Anshus and Ruckenstein (1974). The criterion for instability is then that $a$ is greater than zero. In the limit of very small Reynolds number, ($Re \rightarrow 0$), for nonzero $\zeta$, and for a very broad range of small and moderate wave number $\alpha$, Anshus and Ruckenstein (1974) obtained

$$a = \frac{1}{3} \zeta Re^{-2/3} = \frac{\sigma}{\rho g h^2 \sin\beta} \quad (9)$$

The result is independent of $\beta$ (if $\cot\beta$ is not extremely large) and the approximations all hold if $Re$ is small compared to unity and, at the same time, $\zeta Re^{-2/3}$ is large compared to unity. According to Equation (9), $a$ is positive and the film is more unstable as the Reynolds number gets smaller. In an extension of this calculation to nonvanishing Reynolds numbers, Anshus and Ruckenstein (1974) have shown that for sufficiently large values of the surface tension parameter $\zeta$ the critical Reynolds number below which the growth parameter $a$ is positive is approximately 4. Simon and Hsu (1970) have found that a continuous wetted surface of water on unheated conducting glass could be maintained only if the Reynolds number is greater than about 13. The agreement of this number with theory is quite satisfying qualitatively. The type of perturbation used in the theoretical treatment describes, in fact, the roping perturbation observed experimentally to generate dry patches. An additional feature of this instability is that the growth parameter $a$ is independent of the cross-stream wave number $\alpha$ at least if $\alpha$ is much smaller than $a$. The significance of this is that one expects essentially all spacings of dry patches to be equally probable. This appears to be an agreement with experimental evidence since dry patches occur with no regular spacing.

*Case iii* $m_{imag} = 0$, $\gamma = 0$, $\cot\beta < 0$, *underside ribbing.* Gerstmann and Griffith (1967) studied experimentally the condensation on the underside of inclined surfaces. As condensate collects on the surface, it begins to flow due to gravity. They observed longitudinal ridges, developing in the flow direction from the crests of which eventually drops fall. Gerstmann and Griffith reported that for steeply inclined surfaces no longitudinal ridges appear. Instead, they observed roll waves which probably masked the ridges. A qualitative experiment carried out by us concerning film draining from the underside of an inclined plate has shown that if the fluid is sufficiently viscous, ridges will appear even on very steep (but nonvertical) plates.

In contrast to the case of dry patches which result from a surface tension driven instability, underside ribs which result from a gravity driven instability occur not only for very thin films but also for thick ones. Nevertheless, for ease of analysis it is assumed that the Reynolds number is very small. In the case of either condensation or draining a stream-wise spacially growing transversally periodic perturbation is observed. Thus the present case can be treated as a steady roping instability. Near neutral stability the stream-wise growth parameter $a$ is small in magnitude compared to the wave number $\alpha$. In these conditions, Equations (4) to (7) reduce to

$$(D^2 - \alpha^2)^2 \hat{v} = 0 \quad (10)$$

$$D\hat{v} = \hat{v} = 0 \quad \text{at} \quad y = 1 \quad (11)$$

$$(D^2 + \alpha^2 - 2)\hat{v} = 0 \quad \text{at} \quad y = 0 \quad (12)$$

$$(D^2 - 3\alpha^2) D\hat{v} + \frac{2\alpha^2}{3a} (3\cot\beta + \alpha^2 \zeta Re^{-2/3}) \hat{v} = 0 \quad \text{at} \quad y = 0 \quad (13)$$

The solution to this set of equations is straightforward and, with no further approximations, results in

$$a = -\frac{\cot\beta}{\alpha} \left( 1 + \frac{\alpha^2 \zeta Re^{-2/3}}{3\cot\beta} \right) \frac{\sinh\alpha \cosh\alpha - \alpha}{\cosh^2\alpha + \alpha^2} \quad (14)$$

In this case as in Case i, there is a finite range of $\alpha$ to which the flow is unstable. That is, $a$ is nonnegative if

$$\alpha^2 \leq -\frac{3\cot\beta}{\zeta} Re^{2/3} \quad (15)$$

Also, as in the case of wave motion and in contrast to the case of dry patches there is a dominant wavelength for which $a$ assumes a maximum. The implication of this is that one can anticipate a vertical rib spacing given by this wave number of maximum growth, a conclusion which is in qualitative agreement with the experimental observations.

## CONCLUSIONS

Three distinct types of instability on falling films have been examined. In the first case, time growing traveling waves on a vertical film occur even down to vanishing Reynolds number, but the time growth of these waves becomes weaker and more limited to long waves as the Reynolds number decreases. In underside film flow, time steady stream-wise growing longitudinal ribs develop at all flow rates, but the spatial growth becomes weaker as the flow rate vanishes. In contrast, the time steady spacially growing instability leading to dry patches appears only below a critical Reynolds number and becomes stronger as the Reynolds number decreases.

## NOTATION

$a$ = nondimensional growth coefficient case (ii)
$c$ = ratio between wave velocity and $\overline{U}_a$
$D$ = differentiation with respect to $y$
$g$ = gravitational acceleration
$h$ = film thickness for the unperturbed case
$m, n$ = space coefficient, Equation (3)
$P$ = pressure
$Re$ = Reynolds number = $\overline{U}_a h/\nu \equiv (gh^3 \sin\beta)/3\nu^2$
$U_x, U_y, U_z$ = velocity components in $x$, $y$ and $z$ direction
$\overline{U}$ = $X$ component of velocity in Nusselt's flow
$\overline{U}_a$ = average velocity of the liquid = $(gh^2 \sin\beta)/3\nu$
$u$ = $U_x/\overline{U}_a$
$\overline{u}$ = $\overline{U}/\overline{U}_a$
$v$ = $U_y/\overline{U}_a$
$X, Y, Z$ = stream-wise, depth, and transverse spatial coordinates
$x, y, z$ = nondimensional spatial coordinates $X/h$, $Y/h$, $Z/h$

### Greek Letters

$\alpha$ = stream-wise (case i) or transversal case (ii) wave number
$\beta$ = angle of plane from horizontal
$\gamma$ = exponent in the expression of the disturbance
$\eta'$ = surface perturbation
$\nu$ = kinematic viscosity
$\rho$ = density
$\sigma$ = surface tension
$\zeta$ = surface tension group = $\sigma(3/g\rho^3\nu^4 \sin\beta)^{1/3}$
$'$ (prime) denotes infinitesimal perturbation quantity
$\wedge$ denotes $y$ dependence of a quantity

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*Manuscript received August 14, 1974; revision received October 9 and accepted October 10, 1974.*


# Diffusion of Grouped Multicomponent Mixtures in Uniform and Nonuniform Media


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Many biological and manmade processes involve diffusion of several distinct chemical species. However, it is often inconvenient or impossible to measure experimentally the concentration of each individual species. In such situations a practice sometimes called *grouping*, *lumping*, or *aggregation* is commonly employed: this involves representation of the mixture by one or more grouped or pseudocomponents which are in fact themselves mixtures. An example of this approach is the grouping together of many compounds into the single pseudocomponent "blood urea nitrogen."

In addition to accommodating experimental difficulties in resolving individual species, use of pseudocomponents reduces the number of variables necessary in the diffusion model. While such model simplification is obviously a laudable outcome, it is important to remember that a cost is also incurred. Models cast in terms of grouped species are rarely exact, as has been shown by several recent studies on the related problem of aggregation in reacting systems (see, for example, Wei and Kuo, 1969; Bailey, 1972; Hutchinson and Luss, 1970; Luss and Hutchinson, 1971; Golikeri and Luss, 1972).

The objective of this work is to explore the impact of lumping on the accuracy of models for multicomponent diffusion in plane sheets, cylinders, and spheres. In particular, it will be shown that use of an average diffusivity obtained from steady state diffusion through a plane sheet usually results in an overestimate of the unsteady diffusion rate for all the geometries mentioned. A similar conclusion is proven for diffusion of a single species through a nonuniform plane sheet. Also, guidelines for choice of grouped species are presented. Both the analysis concept and the particular mathematical devices used here derive directly from the work of Hutchinson and Luss (1970). The following presentation is limited to situations where the transport of each component can be described by the Fick's Law form with constant diffusion coefficient.

## DIFFUSION THROUGH A PLANE SHEET

The first diffusion medium to be considered is a homogeneous plane sheet of thickness $L$ which at time zero