

NOTATION

C_T	= total concentration, gm.-moles/cc.
D_{AB}	= bulk diffusivity of substance A in substance B, sq.cm./sec.
D_K	= Knudsen diffusivity of substance A, sq.cm./sec.
F	= molar diffusion rate of substance A, gm.-moles/sec.
l	= total of length of given size subpore within a single pore assembly, cm.
L	= l_I/l_{II} , dimensionless
m	= number of cells in a single pore assembly, dimensionless
n	= number of pore assemblies within the porous pellet, dimensionless
r	= radius of pore, cm.
R	= r_I/r_{II} , dimensionless
y	= mole fraction of substance A, dimensionless
α	= $1 + (\text{ratio of molar rates of diffusing substances})$, dimensionless
γ	= constriction factor—contribution of pore constrictions to tortuosity factor, dimensionless
I	= property of pore of radius r_I
II	= property of pore of radius r_{II}
B	= bulk diffusion completely controlling
K	= Knudsen flow completely controlling

"	= predicted in situation where present method of obtaining pore-size distributions is used
'	= predicted in situation where true pore-size distribution is used

LITERATURE CITED

1. Brown, L. F., H. W. Haynes, Jr., and W. H. Manogue, *J. Catalysis*, **14**, 220 (1969).
2. Foster, R. M., and J. B. Butt, *AIChE J.*, **12**, 180 (1966).
3. Haynes, H. W., Jr., Ph.D. thesis, Univ. Colorado, Boulder (1969).
4. Johnson, M. F. L., and W. E. Stewart, *J. Catalysis*, **4**, 248 (1965).
5. McBain, J. W., *J. Am. Chem. Soc.*, **57**, 699 (1935).
6. Michaels, A. S., *AIChE J.*, **5**, 270 (1959).
7. Satterfield, C. N., and P. J. Cadle, *Ind. Eng. Chem. Fundamentals*, **7**, 202 (1968).
8. Wakao, N., and J. M. Smith, *Chem. Eng. Sci.*, **17**, 825 (1962).
9. Walker, P. L., Jr., F. Rusinko, Jr., and E. Raats, *J. Phys. Chem.*, **59**, 245 (1955).
10. Wheeler, A., in "Advances in Catalysis," p. 249, vol. III, W. G. Frankenburg et al., ed., Academic Press, New York (1951).
11. Wicke, E., and R. Kallenback, *Kolloid-Z.*, **97**, 135 (1941).

Bimodal Wave Formation on Thin Liquid Films Flowing down a Plane

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In a recent paper Krantz and Goren (8) presented the first measurements of wave lengths, wave velocities, and amplification rates of waves on thin liquid films flowing down an inclined plane, resulting from imposed disturbances of controlled amplitude and frequency. In a second paper the authors (9) presented measurements of equilibrium wave amplitudes for viscous oils and compared these data to a new theory describing finite-amplitude, two-dimensional, equilibrium waves on thin films flowing down an inclined plane.

This note describes the formation of "bimodal waves" on thin liquid films flowing down an inclined plane. A "bimodal wave" consists of a fundamental wave and a second superimposed distinct wave having the first harmonic frequency of the fundamental wave. In general these component waves of the bimodal wave have different wave velocities and amplification rates and thus are observed to pass each other and change form as they travel down the column. Such waves also were reported by Tailby and Portalski (12), Jones (5), and Hallett (4) in their studies of thin liquid films subject to room disturbances. It is important to note that bimodal waves do not imply a Fourier decomposition of a nonsinusoidal wave form. Bimodal waves refer to two distinct wave forms traveling down the column with distinct wave velocities.

LINEAR STABILITY THEORY

Attempts to describe the formation and growth of waves on thin liquid films can be broadly classified into linear and nonlinear stability theories (7 to 9). Investigations

into the theory of linear stability are based on the assumption that infinitesimally small disturbances are superimposed on the laminar film flow. The theory has been developed in an effort to understand the mechanism and conditions necessary for transition away from unrippled laminar film flow and, as such, can predict only exponential growth or decay of infinitesimal disturbances.

The next step in the understanding of the transition away from unrippled laminar film flow is the consideration of disturbances which have grown too large to be governed by a theory of infinitesimal disturbances. In order to predict the behavior of these larger amplitude disturbances and to understand the physical processes which must occur to slow down the exponential growth it is necessary to consider the nonlinear terms in the equations of motion.

One conclusion which can be drawn from the linear stability theory of thin films flowing down a plane is that the flow becomes unstable at very small Reynolds numbers; vertical film flow is always unstable. Hence, at these small Reynolds numbers viscous forces are of comparable magnitude to gravity and inertia forces. Very few theories of finite amplitude waves have appeared which consider viscous effects based on rigorous hydrodynamics.

NONLINEAR STABILITY THEORY

With the exception of the equilibrium wave amplitude correlation developed by Krantz and Goren (9), all the nonlinear theories, which include both viscous terms and surface tension (1, 3, 6, 10, 11), consider only small amplitude waves. These theories thus are restricted to

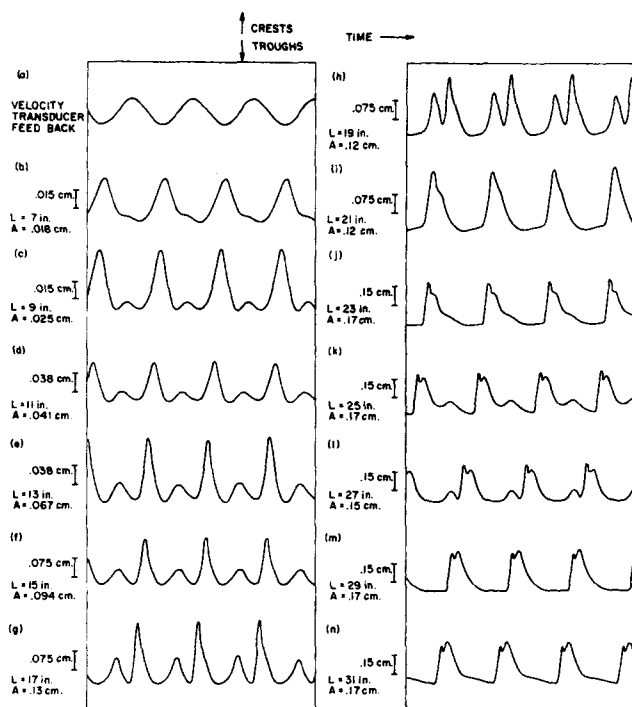


Fig. 1. Wave traces for bimodal waves: $N_{Re} = 0.92$, $N_L = 9.4$, $\beta = 74.5^\circ$, $\alpha = 0.12$.

describing nearly sinusoidal waves because including terms other than the first term in the Fourier integral representation of the wave will introduce higher order terms in the wave amplitude. Thus the nonlinear effects being considered by most theories are the finite dissipation of energy by the waves and the drain of energy from the mean flow by the growing waves; the nonlinear inertial transfer of disturbance energy is not considered. Neglecting this nonlinear effect precludes the possibility of predicting the appearance and behavior of bimodal waves.

Several nonlinear stability theories (1, 6, 11) predict that the transition away from unrippled laminar film flow will lead to a second laminar flow consisting of two-dimensional waves at their equilibrium amplitude. This equilibrium amplitude corresponds to an exact balance between the rate at which the waves extract energy from the mean flow and the rate at which they dissipate energy. Of course, this second equilibrium amplitude laminar flow may in turn be hydrodynamically unstable. However, no stability analysis has been attempted for the equilibrium amplitude wave flow.

The observation of bimodal waves is of significance in that they represent the first step by which an unstable flow can build up a nearly continuous spectrum of superimposed two-dimensional disturbances from some fundamental disturbance (which usually is the most highly amplified wave for that flow). Whether or not this build-up of a spectrum of superimposed waves will occur depends on the hydrodynamic stability of the equilibrium amplitude flow of the fundamental wave. Neither the occurrence of bimodal waves nor the stability of the equilibrium amplitude flow of the fundamental wave has been considered in the nonlinear wave theories developed to date. Thus the results described here indicate areas wherein our research effort on thin liquid films should be directed.

EXPERIMENTAL DESIGN

The apparatus and experimental procedure have been de-

scribed elsewhere (7, 8). Briefly, a plane wetted-wall column was used. Waves of controlled amplitude and frequency were imposed on the flow by a thin wire inserted into the flow and vibrated normally to the flow surface. Wave amplitudes were measured by a light extinction technique with a photoconductive cell. The fluids used were Chevron No. 5 white oil ($\nu = 0.499$ poise, $\rho = 0.853$ g./cc., $\sigma = 29.5$ dynes/cm. at $25^\circ\text{C}.$) and Chevron No. 15 white oil ($\nu = 1.46$ poise, $\rho = 0.868$ g./cc., $\sigma = 30.8$ dynes/cm. at $25^\circ\text{C}.$).

The fluctuating resistance of the photoconductive cell was converted into a voltage signal and displayed on one channel of a dual-channel recorder; the second channel displayed the velocity transducer feedback from the vibration generator used to impress the disturbance on the flow. With the extinction coefficient of the dyed oil known, the wave amplitude and frequency were determined from the recorder traces. Wave velocities were obtained by visually following a wave crest and timing its passage between two points separated by a known distance. Alternately the wave velocity could be obtained by observing the Lissajous figures produced on an oscilloscope when the input to the vibration generator was plotted against the measured wave form signal.

RESULTS AND DISCUSSION

Figure 1 gives wave patterns obtained for a 3.0 cps. imposed vibration at Reynolds number, $N_{Re} = 0.92$, surface tension parameter, $N_L = 9.4$, column angle, $\beta = 74.5^\circ$, wave number, $\alpha = 0.12$, and film thickness, $h_0 = 0.10$ cm. Panel (a) shows the feedback from the velocity transducer. The traces have been arranged so that the signals are synchronous because the origin for each has been chosen at the same phase point of the input disturbance; that is, each trace begins at the same instant in time. In panel (b), 7 in. downstream from the vibration generator, the fundamental wave has an amplitude of 0.018 cm. and a frequency of 3.0 cps. A small secondary wave is seen emerging from the trailing edge of the fundamental wave. Growth of the fundamental and secondary waves is shown in subsequent panels. (Note that the amplitude scale is not the same for all panels.)

One significant observation in the studies of Krantz and Goren (8) on the growth or decay of unimodal two-dimensional waves was that the wave length and velocity remained constant with distance downstream even when the wave amplitude approached the equilibrium amplitude. Hence, linear stability theory can predict the wave length and velocity of both infinitesimal and finite amplitude unimodal waves. One might hope that it could do equally well for the component waves of the bimodal waves.

The experimentally determined wave velocities of 16.7 cm./sec. for the 3.0 cps. fundamental and 15.2 cm./sec. for the 6.0 cps. secondary wave compare favorably with the 16.3 cm./sec. and 15.5 cm./sec. predicted by the modified Orr-Sommerfeld equation of Anshus and Goren (2), assuming that each wave grows independently according to linear stability theory. This close agreement with the predictions of linear stability theory is quite interesting in view of the fact that the secondary wave arises from the nonlinear inertia terms in the equations of motion.

At 20 in. from the disturbance generator, the secondary wave attained an amplitude nearly equal to that of the fundamental. Somewhere between 20 and 21 in. [panel (i)], the fundamental wave finally caught up to the secondary wave immediately in front of it. Surprisingly, the secondary wave which was seen emerging from the trailing edge of the fundamental at the next column position [panel (j)] was considerably reduced in amplitude. At this position the fundamental wave had reached its two-dimensional equilibrium amplitude, $A = 0.17$ cm. The apparent splitting of the fundamental peak in panel

(f) is a distortion of the true wave profile due to refraction of the light beam. This refraction effect was slightly noticeable already in panel (g). However, it should not affect the determination of the wave amplitude, for no refraction occurs at wave peaks or troughs. As the waves progressed further down the column, the amplitude of the fundamental waves remained essentially constant whereas the secondary wave ultimately completely decayed away. It thus appears that the two-dimensional equilibrium amplitude flow for a 3.0 cps. wave is stable with respect to the secondary wave, even though the original unrippled laminar flow was found to be unstable when subjected to a 6.0 cps. vibration.

No secondary waves were observed for impressed fundamental waves of higher frequency for these flow conditions. However, when lower frequency fundamental waves were impressed upon the flow, multiple frequency secondary waves were observed. These secondary waves appeared to have frequencies which were integral multiples of that of the impressed fundamental wave. The ability to observe bimodal waves was quite dependent on the amplitude of the impressed fundamental wave. Given a low enough frequency, bimodal waves were more likely to occur for larger amplitudes of the impressed fundamental wave.

That the equilibrium amplitude flow was stable with respect to the secondary wave generated by the flow was not always the case. The ultimate nature of the flow at the downstream end of the column depended on the particular flow conditions and the amplitude and frequency of the fundamental wave being impressed on the flow. More often than not, the flow became unstable with respect to three-dimensional disturbances and a completely chaotic pattern of waves resulted.

The appearance of the bimodal waves on the column suggested an experiment wherein multiple frequency waves were impressed on the flow. With separate control of the amplitude and frequency of each disturbance, nearly any combination of waves could be studied subject to the same limitations as were encountered with single wave generation. These studies were not restricted to having one frequency an integral multiple of the other. Wave forms similar to those obtained for bimodal wave generation could be observed when an unstable wave and its first harmonic were simultaneously impressed on the flow.

Observations on the occurrence of bimodal waves are summarized in Figure 2. This is a typical amplification curve predicted by linear stability theory for an unstable flow showing the amplification factor αc_i as a function of wave number α . The linear stability theory development

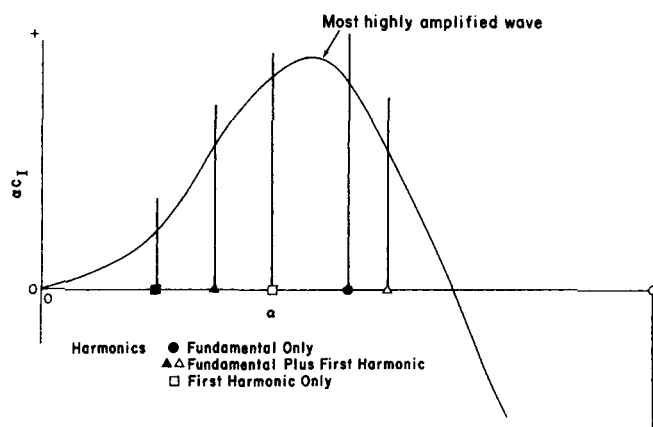


Fig. 2. A typical amplification curve.

leading to the amplification factor predictions is discussed elsewhere (2, 7, 8). A positive amplification factor corresponds to an unstable wave whereas a negative amplification factor corresponds to a stable wave. If the first harmonic of the impressed fundamental is much more highly amplified than the fundamental, then only the first harmonic is observed. If the fundamental and its first harmonic have comparable amplification rates, both waves (bimodal waves) typical of those shown in Figure 1 would be observed. If the first harmonic is a stable wave, then only the fundamental would be observed.

CONCLUSIONS

Bimodal waves were observed on low Reynolds number film flow when the first harmonic of the unstable wave being impressed on the flow was an unstable wave of comparable growth rate. The bimodal wave was composed of two waves whose wave number and wave velocity were predicted quite well by linear stability theory. The equilibrium amplitude flow of the unstable wave which was impressed on the flow appeared under certain flow conditions to be stable with respect to the wave number of the secondary wave composing the bimodal wave. That is, the secondary wave ultimately decayed away once the fundamental wave reached its two-dimensional equilibrium amplitude.

ACKNOWLEDGMENT

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NOTATION

A	= wave amplitude, one-half peak-to-peak
c_i	= imaginary part of complex dimensionless wave velocity
g	= acceleration of gravity
h_0	= mean film thickness
L	= streamwise distance from point of wave incipitation
N_{Re}	= $\bar{u}h_0/\nu$, Reynolds number
N_i	= $\sigma 3^{1/3}/\rho g^{1/3} \nu^{4/3}$
\bar{u}	= mean velocity
α	= $2\pi h_0/\lambda$, wave number
β	= angle of column to horizontal
λ	= wave length
ν	= kinematic viscosity
ρ	= density
σ	= interfacial tension

LITERATURE CITED

1. Anshus, B. E., Ph.D. thesis, Univ. California, Berkeley (1965).
2. Anshus, B. D., and S. L. Goren, *AIChE J.*, **12**, 1004 (1966).
3. Benney, D. J., *J. Math. Phys.*, **45**, 150 (1966).
4. Hallett, V. A., *Intern. J. Heat Mass Transfer*, **9**, 283 (1966).
5. Jones, L. O., M. S. thesis, Northwestern Univ., Chicago (1965).
6. Kapitza, P. L., *Zh. Eksperim. i Teor. Fiz.*, **18**, 3 (1948); English translation in "Collected Papers of P. L. Kapitza," MacMillan (1964).
7. Krantz, W. B., Ph.D. thesis, Univ. California, Berkeley (1968).
8. Krantz, W. B., and S. L. Goren, *Ind. Eng. Chem. Fundamentals*, in press (1970).
9. Krantz, W. B., and S. L. Goren, *Ind. Eng. Chem. Fundamentals*, **9**, 107 (1970).
10. Mei, C. C., *J. Math. Phys.*, **45**, 266 (1966).
11. Nakaya, C., and R. Takaki, *J. Phys. Soc. Japan*, **23**, 638 (1967).
12. Tailby, S. R., and S. Portalski, *Trans. Inst. Chem. Engrs. (London)*, **40**, 114 (1962).