LIQUID FILMS IN VISCOUS FLOW

A trace of radioactive material dissolved in a liquid enables the thickness of a moving film to be determined by measurement of the radiation emitted. The method is rapid and accurate and an average thickness is obtained directly even though the surface of the film may be irregular. Six liquids, having viscosities ranging from 0.5 to 20 centipoises were observed in flow down the inner wall of a vertical tube. Liquids having viscosities of about 1 centipoise or less exhibit typical viscous behavior with respect to film thickness even when waves are present. Liquids having larger viscosities give values of the film thickness which are less than for true viscous flow. The departure from normal behavior increases with increasing viscosity and occurs only over the region where the liquid moves in wave flow. Surface tension is not a factor in either wave formation or wave flow.

The wave motion appears at flow rates well within the viscous region and occurs when the Froude number exceeds unity. Equations derived for the viscous flow of liquid films on the inner wall of a vertical tube would be required where tube diameter is small or liquid viscosity large. Ordinarily the less complex equations for flow down a flat plate may be used. A theory of flow in the viscous region with waves present is suggested.

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Liquid films are encountered in some types of heat and mass transfer equipment, both industrial and experimental, and are of general interest to hydrodynamical theory. The importance of the mechanics of film flow to the analysis of performance of industrial equipment has recently been discussed (*4*). Several different approaches have been employed in measuring film thicknesses, a procedure that is complicated by the fact that the liquid may assume a wave motion at flow rates well within the viscous flow region.

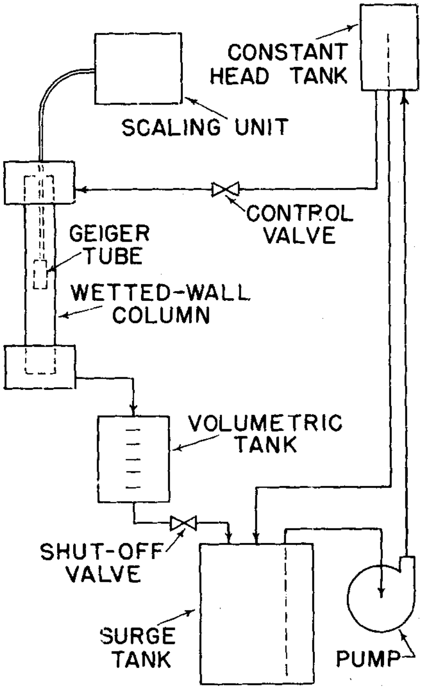

FIG. 1. FLOW DIAGRAM FOR THE MEASUREMENT OF FILM THICKNESS BY USE OF RADIOACTIVE ISOTOPES.

Kirkbride (*12*) made direct measurements with a micrometer and reported that the thickness at the wave peak was several times the thickness of the film expected for normal laminar flow. Others (*3, 5*) obtained average film thickness by measuring drainage after stopping the flow of liquid simultaneously at the top and bottom of a vertical tube. This method gave satisfactory results at moderate flow rates but was subject to timing errors at high rates. It was reported that in spite of the considerable height of the wave crests, the flow behavior was that expected for true viscous flow. However, recent work has shown that when waves are present the apparent surface velocity is greater than normal (*6, 10*), and for the mass transfer process in a wetted-wall column (*9, 11, 16*) liquid-film resistances are negligibly small, possibly because of a mixing action occurring in the film.

Heat transfer to and through liquid films on column walls has been observed, particularly in the case of condensing vapors (*2*), and the heat transfer rate is above that which would be expected from viscous theory. A portion of the viscous region for flow with a free surface thus exhibits an anomalous behavior and additional information was desired. The use of radioactive tracers offers an excellent method of observing film thickness without disturbing the flow pattern.

METHOD OF USING RADIOACTIVE TRACERS

A trace of a radioactive isotope is dissolved in a liquid which is made to flow as a film down the inner wall of a vertical tube. A Geiger-Mueller tube positioned at the center of the vertical tube registers the radioactivity emitted by the liquid. The activity observed depends on the thickness of the liquid film. Samples of the liquid must be analyzed for the concentration of radioactivity because absorption may cause changes in

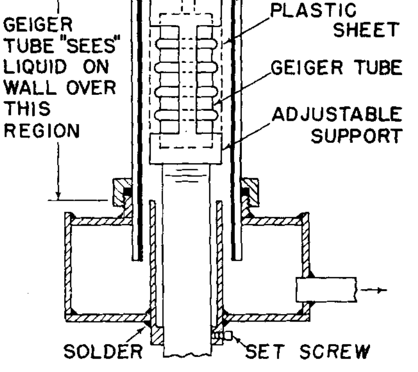

FIG. 2. DETAILS OF COLUMN CONSTRUCTION AND GEIGER-TUBE PLACEMENT.

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concentration during the course of a run. Also, because of the trace quantities employed, it is not feasible to introduce the same concentration of activity into each liquid.

Equations Relating Radioactivity and Film Thickness. The isotope used was yttrium 91, which emits only beta radiation (1.53 mev.) and has a half life of 57 days. A linear relation does not exist between observed activity and thickness of the liquid film because beta radiation is partially absorbed by the liquid. However, corrections may be made and all observations reduced to a common basis by the following expressions:

$$m = C \times A_c \qquad (1)$$

$$A_c = A_o P_a P_c \qquad (2)$$

The term A_o is the activity registered by the scaling unit for flow of the liquid film down the vertical tube. The factor P_a corrects for the adsorption of the liquid film and is the ratio of the activity that the film would show without absorption to the activity observed with absorption. The factor P_c corrects the observed activity for the concentration of isotope. It is the ratio of the activity of a liquid sample to the activity of some liquid selected arbitrarily as a basis. These factors are determined by analytical methods described later. The activity of a liquid sample on the date of analysis (A) must be corrected for radioactive decay to the activity the sample had on the date of its evaluation in the flow apparatus (A'). The following expression is used for this:

$$A/A' = e^{-\lambda t} \qquad (3)$$

For Y^{91}, $A/A' = 1/2$ at $t = 57$ days and $\lambda = 0.01216$ (day)$^{-1}$. The term A_c is therefore the activity that the counting apparatus would register if each liquid had zero absorption and the same concentration of radioactive isotope. The constant C is the proportionality factor between corrected activity and the film thickness. It depends on the geometry and counting efficiency of the Geiger tube and is determined by operating the flow equipment in a region where the film thickness can be calculated from viscous theory. Thus a film thickness determined by the use of a radioactive tracer is not measured directly but is computed from the ratio of an unknown to a known condition of flow.

Equations of Flow. Equations for the viscous flow of a liquid film down a vertical flat plate have been established(5) and are given below:

$$m = \left[\frac{3\ \mu\ Q}{g_c\ \rho_a}\right]^{1/3} \qquad (4)$$

$$v = \frac{\rho_a\ g_c}{\mu}\left(mx - \frac{x^2}{2}\right)^{1/2} \qquad (5)$$

$$V_s = \frac{\rho_a\ g_c\ m^2}{2\mu} \qquad (6)$$

$$V = \frac{\rho_a\ g_c\ m^2}{3\mu} \qquad (7)$$

$$V_s/V = 1.5 \qquad (8)$$

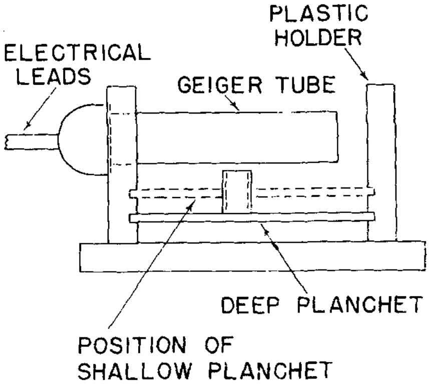

FIG. 3. ARRANGEMENT OF PLANCHETS AND GEIGER TUBE FOR THE DETERMINATION OF SAMPLE ACTIVITY.

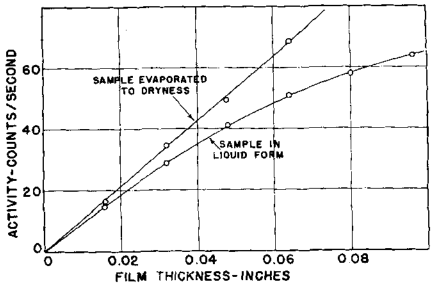

FIG. 4. TYPICAL DATA FOR THE SELF-ABSORPTION OF BETA RADIATION BY A LIQUID (70.4% GLYCEROL SOLUTION).

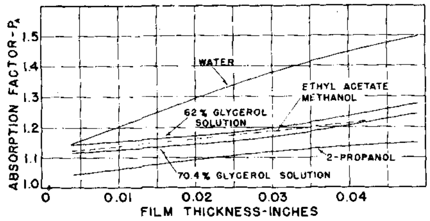

FIG. 5. DEPENDENCE OF THE ABSORPTION FACTOR P_A ON FILM THICKNESS.

EXPERIMENTAL EQUIPMENT AND PROCEDURE

Apparatus. The equipment consisted of a device for rigidly supporting a removable glass tube in a vertical position and for supplying the inner wall of the tube with liquid. This equipment is represented diagrammatically in Figure 1. Liquid at a constant rate was provided by a constant head tank. Glass, copper, and copper alloys were used throughout to prevent contamination by solid corrosion products which would absorb radioactive ions and deposit in the wetted-wall tube. Liquid-flow rate was measured by means of a calibrated glass vessel inserted in the drainage line between the vertical tube and the surge tank. This method gave accurate results for extreme ranges of flow rate and viscosity.

The vertical glass tube had an inside diameter of 1.423 ± 0.010 in. The upper end of the glass tube was ground to an angle of 90° with the walls and the upper edge of the inside wall was roughened slightly to ensure uniform wetting. The tube was brought into vertical alignment by use of a precision square and level. Any deviation from the vertical was of the same order of magnitude as the variation in straightness of the glass walls. The tube could be readily removed for cleaning, as was done prior to each run to reduce the background count to a minimum. Before overflowing the top of the glass tube, liquid entered an enlarged cylindrical well containing two screens concentric with the tube. No liquid disturbance in this well was apparent except at the very highest flow rates used.

Figure 2 shows the column construction details. The Geiger-Mueller tube for determining the activity of the liquid flowing on the walls was a Victoreen Model 1B85 and was held concentric with the glass tube. The holder shielded the Geiger tube to limit the area it could "see" to the immediate vicinity. This eliminated the influence of radiation from the upper and lower portions of the glass tube, where the flow pattern is not typical. The top of the Geiger tube was 12 in. below the top of the glass tube.

Procedure. A Nuclear model 165 scaler supplying 850 volts to the Geiger-Mueller tube was used for counting. The same counting tube and apparatus were used for all determinations. In most cases the number of counts obtained was 10,000 or more, which gave a standard deviation of 100 or less. The flow rate was established simultaneously with the counting period. The background employed was that at the beginning of a run since some residual activity remained after a run due to liquid holdup and subsequent evaporation. The entire flow range was covered several times during a series of determinations to ensure that the data were representative.

The isotope Y^{91} was received as YCl_3 in 6N HCl acid. A stock solution of 2 mcuries of Y^{91} in 8 liters of isopropyl alcohol was prepared. Portions of this (500 to 2,000 ml.) were evaporated to a solid residue, which was then taken up by a portion of the liquid under test. In the case of methanol and ethyl acetate the residue was insoluble, and for these a portion of the original Y^{91} acid solution was placed directly in the liquid. For each flow determination 8 liters of liquid was used and 2g. of anhydrous aluminum chloride was added as a carrier to reduce loss of radioactive isotope by adsorption on the walls of the apparatus.

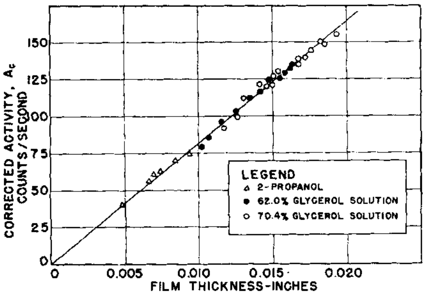

FIG. 6. CORRECTED ACTIVITY VS. CALCULATED THICKNESS FOR FLOW WITHOUT WAVES.

Because evaporation and adsorption may cause the activity to change during the course of a run, and because a solution may not easily be prepared to have a predetermined concentration of isotope, a method of analysis of liquid activity was necessary. As dip counting was not satisfactory, a method using planchets was developed. The planchets were constructed of short sections of 1/2-in. copper tubing held to the backing plate by solder, litharge-glycerin, or wax depending on the liquid under test and the type of backing used.

For comparing samples of the same liquid a deep planchet was used, 2 ml. of liquid being added to the planchet and the activity determined by the apparatus shown in Figure 3. For comparing the activity of different liquids, 0.100 ml. of the sample was placed in a shallow planchet and was then evaporated to dryness before counting. This method could have been used for all samples, but the process of analysis was speeded considerably by using the deep planchet for a series of samples of the same liquid, and the evaporative technique for only one sample of each series. The procedure is further illustrated by the sample calculation given later.

To establish values for the factor P_a it was necessary to determine the absorption of beta radiation as a function of film thickness. For this, shallow planchets having a backing plate of glass were used. This was necessary to simulate the conditions of the liquid on the wall of the glass tube because the use of a copper backing material gave results appreciably different from those for the glass. Small increments of liquid were added by micropipet and the corresponding activity was measured. Film thickness was calculated from liquid volume and planchet diameter. Surface-tension effects were reduced in the case of water and glycerol solutions by adding a small quantity of a surface-active agent. Activity values at zero absorption were obtained by evaporation. A typical absorption curve is shown in Figure 4, and values of the absorption factor as a function of film thickness are given in Figure 5.

TABLE 1.—WAVE INCEPTION AS A FUNCTION OF COLUMN POSITION AND REYNOLDS NUMBER

Liquid (in order of increasing viscosity)	Distance below top of wetted-wall column, in.			
	0*	4	8	12†
Ethyl acetate	<66	880	1,670	
Methanol	<60	760	1,470	
Water	15	530	1,030	1,540
2-Propanol	15	340	660	970
62.0% Glycerol solution	18	180	330	
70.4% Glycerol solution	12	130	230	

*The distance for initial appearance of the waves was small and was assumed zero.
†This distance coincided with the top of the Geiger tube.

Materials. Six different liquids were employed in the test apparatus: ethyl acetate, methanol, water, 2-propanol, 62.0% glycerol solution, and 70.4% glycerol solution. These provided a viscosity range of 0.5 to 20 centipoises. The following grades of materials were employed: distilled water, U.S.P. glycerol, Shell 2-propanol (98-99%), Carbide and Carbon Corporation commercial-grade ethyl acetate (87-89%), which analyzed 88.0%, and methanol of unknown source. The glycerol solutions were analyzed by density measurements (*17*) and viscosities were determined from compositions (*21*). Experimental measurements of viscosity for 2-propanol, ethyl acetate, and methanol were made by use of an Ostwald drip-type viscosimeter (Fisher). Values obtained checked handbook values closely and the methanol was indicated to be of high purity. For some of the water and glycerol series, 0.1% by weight of Ethomeen C/15 (Armour) was added to reduce the surface tension of these liquids to more than one half the normal values and provide an excellent method of observing surface-tension effects without change in kinematic viscosity. It was also necessary to add about 0.1% octyl and n-butyl alcohols to reduce foaming.

EXPERIMENTAL RESULTS

It was necessary first to establish the value of the constant, C, in Equation (1). This was done by operating the vertical-tube apparatus under conditions of flow where film thickness could be calculated from flow rate using Equation (4). This may be done for the region where the flow is truly viscous and waves are absent. It was not possible to secure uniform coverage of the wall at the low flow rates required except for the more vicious liquids. The value of the constant was determined by plotting corrected activity vs. calculated thickness, as shown in Figure 6. The best line though the data points and the origin gives $C = 0.0000\text{-}1010$.

Calculations. A sample calculation will serve most conveniently to illustrate the method. The concentration of activity for a sample of I-52 (70.4% glycerol series) as determined by the deep planchet method for example, was 5.27 counts/sec. compared to 5.17 ct./sec. for a sample of I-8. The latter was selected as the sample for the series to be analyzed by evaporation and gave $A = 2.65$ ct./sec. Hence the activity that sample I-52 would have shown on evaporation is

$$A = \frac{5.27}{5.17} \times 2.65 = 2.70 \text{ ct./sec.}$$

These analyses were made 12 days after observations were taken in the flow apparatus, and by Equation (3) the activity corrected to the date of the run is

$$\frac{A}{A'} = e^{-0.01216 \times 12} = 0.864$$

$$A' = 2.70/0.864 = 3.11 \text{ ct./sec.}$$

A sample from the water series, G-2 was selected as the activity basis for all determinations and this sample analyzed $A' = 4.50$ct./sec. The value of the correction factor, P_c, for sample I-52 is therefore $P_c = 4.50/3.11 = 1.450$
This signifies that at the time the flow determination for the glycerol solution was made the concentration of radioactivity was less than that of sample G-2 by this ratio. Hence the activity indicated by the Geiger tube in the flow apparatus for run I-52 ($A_0 = 162.0$ ct./sec.) must be increased by 1.450 so that the two series of determinations may be directly compared. In practice the value of P_c was not determined for each point because it changed only little if at all for most liquids. Four or five samples were taken over a series of determinations, and P_c was interpolated with respect to time.

The factor P_a must be obtained by trial since this depends on the film thickness, which is yet unknown. A first approximation of thickness is obtained from Equation (4) for true viscous flow. For I-52 this is

$$m = \left[\frac{3 \times 0.449 \times 16.29}{4.17 \times 10^8}\right]^{1/3} = 0.00374 \text{ ft. } (0.0449 \text{ in.})$$

From Figure 5, $P_a = 1.23$, which signifies that the flow activity observed would have been greater by this factor if absorption of radiation by the film had not occurred. By Equation (2) the corrected flow activity is

$$A_c = 162.0 \times 1.450 \times 1.23 = 288 \text{ ct./sec.}$$

and by Equation (1) the film thickness is
$m = 0.00001010 \times 288 = 0.00291$ ft. (0.0345 in.)

Since the calculated film thickness does not check the assumed value, a new value of $P_a = 1.19$ is selected corresponding to the calculated thickness. A recalculation gives $m = 0.00282$ ft. (0.0334 in.) and no further refinement in the value of P_a is necessary. It may be noted that large variations in film thickness result in only small changes of the factor P_a. Therefore, the use of this factor based on average thickness results in only a small error even though waves of considerable height are present.

Characteristics of Wave Flow. Flow rates, corresponding to Reynolds numbers of 4 to 5,000, were employed and covered the entire region of viscous flow although it was not possible to obtain this range with each liquid. The surface of the liquid film assumes a wave motion at very low flow rates. For the lighter liquids it was not possible to operate the apparatus in a nonwave region because of incomplete coverage of the wall. The initial formation of the waves was sharply defined and the appearance or disappearance of the waves could be effected by only a

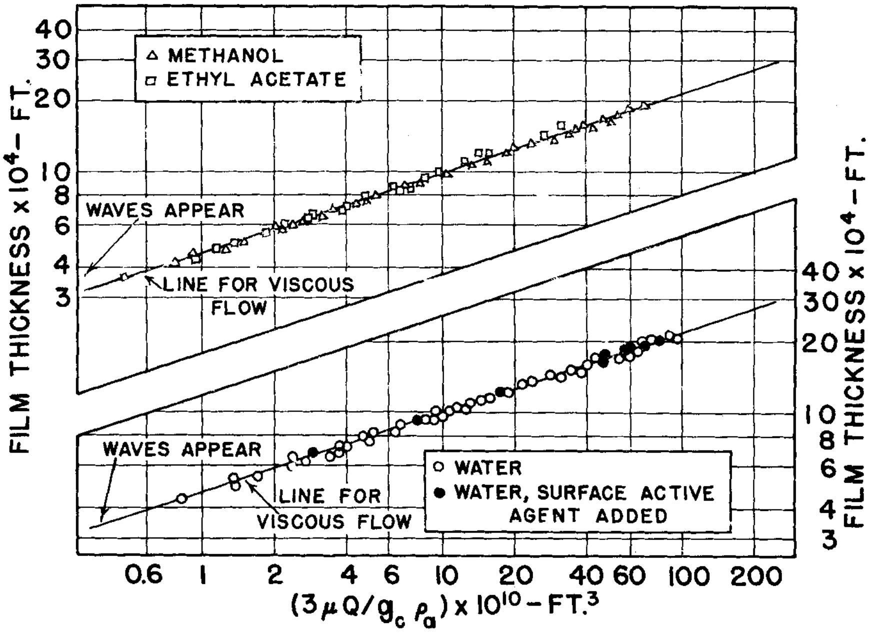

Fig. 7. Film Thickness for Water and Liquids Less Viscous Than Water.

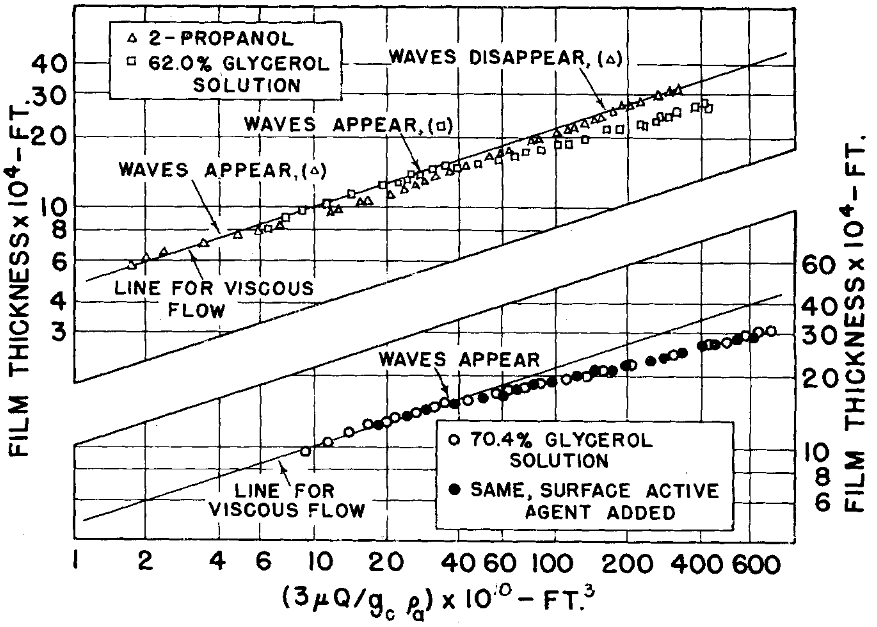

Fig. 8. Film Thickness for Liquids More Viscous Than Water.

small change in the flow rate. At inception the waves appeared over the entire column except for a very short distance at the top. As the flow rate increased, the point of wave formation progressed down the column, evidently because the liquid was accelerating in the upper portion. Table 1 shows this type of behavior and it is seen that a certain distance is required to establish the wave motion. This distance increases linearly with the flow rate over the range observed (the values given in Table 1 were read from the best lines through the experimental points) and increases with the viscosity. The spacing between waves was estimated at 1.25 to 1.50 in. at inception. This spacing decreased as the rate of flow increased and rapidly reached a condition where it could no longer be followed easily with the eye.

TABLE 2.—TYPICAL FLOW AND ACTIVITY DATA

Determination	Q, $\frac{\text{cu. ft.}}{\text{(ft.) (hr.)}}$	Temperature, °C.	μ/ρ_a, sq. ft./hr.	$\left[\frac{3\ \mu\ Q}{g_c\ \rho_a}\right]$ ft.×10^9	A_o, ct./sec.	P_a	P_c	A_c, ct./sec.	m, ft.×10^3	Re
K series	Ethyl acetate									
9	0.338	26.8	0.01940	0.0471	16.2	1.06	2.36	36.2	0.366	66
19	5.29	26.0	0.01948	0.742	31.5	1.09	2.88	88.6	0.895	1,085
1	18.72	26.1	0.01946	2.62	66.1	1.11	2.34	146.9	1.484	3,850
L series	Methanol									
18	0.399	29.0	0.0265	0.0763	37.3	1.11	1.009	41.8	0.422	60
1	15.25	28.8	0.0266	2.92	118.5	1.15	1.009	137.6	1.390	2,240
27	30.3	29.5	0.0264	5.76	155.7	1.16	1.009	182.2	1.840	4,590
G series	Water									
13	0.642	33.0	0.0295	0.1363	26.8	1.16	1.677	52.2	0.527	87
15	21.2	33.0	0.0295	4.52	76.3	1.30	1.722	170.8	1.725	2,880
2	27.8	31.8	0.0302	6.04	88.4	1.30	1.522	174.9	1.767	3,690
G series	Water, surface-active agent added									
40	1.357	33.0	0.0295	0.288	28.8	1.20	1.980	68.5	0.693	184
39	22.2	33.0	0.0295	4.71	68.9	1.30	1.975	177.0	1.788	3,010
47	32.1	32.2	0.0300	6.93	75.0	1.31	2.010	199.5	2.01	4,280
J series	2-Propanol									
17	0.261	28.0	0.0920	0.1724	65.9	1.12	0.774	57.1	0.576	11.3
5	12.13	26.8	0.0950	8.29	220	1.17	0.736	189.5	1.920	512
42	43.7	25.6	0.0982	30.8	305	1.21	0.832	307	3.10	1,780
H series	62.0% Glycerol solution									
13	0.767	32.5	0.0253	1.395	140.6	1.16	0.693	113.1	1.142	12.1
34	6.63	34.0	0.238	11.34	221	1.18	0.693	180.9	1.827	111.3
40	18.08	34.0	0.238	31.0	290	1.20	0.693	241	2.44	304
I series	70.4% Glycerol solution									
24	0.461	34.0	0.415	1.374	68.6	1.14	1.455	113.8	1.149	4.4
7	15.29	32.0	0.453	49.7	159.1	1.19	1.455	275	2.78	142.0
45	25.2	32.7	0.439	79.3	178.5	1.21	1.450	312	3.15	229
I series	70.4% Glycerol solution, surface-active agent added									
78	0.818	32.0	0.453	2.66	80.4	1.14	1.464	134.2	1.356	7.2
56	5.38	32.2	0.449	17.37	124.2	1.17	1.450	211	2.14	47.8
52	16.29	32.2	0.449	52.6	162.0	1.19	1.450	276	2.82	145.0

Film Thickness. Typical data are given in Table 2 for each of the liquids employed. Because of the behavior obtained, the data are conveniently placed into one of two groups according to whether the liquid is more viscous or less viscous than water. Figure 7 shows experimental film thickness plotted against a grouping of terms suggested by Equation (4). For liquids of viscosity equal to water or less, the agreement between observed and theoretical is good even though most of the flow lies in the region where waves are present.

Figure 8 shows the behavior of liquids having viscosities greater than the viscosity of water. Film thickness departs from that for true viscous flow at the point of wave inception and is less than theory predicts. In the case of 2-propanol film thicknesses are observed to return to the expected values when the waves disappear. The magnitude of the difference between experimental and theoretical thickness increases with viscosity for the region of wave flow. Sufficiently high flow rates could not be obtained to observe the behavior of the more viscous liquids near the turbulent region.

Effect of Surface Tension. The activities and flow rates of several liquids were determined before and after the addition of a surface-active agent, which was estimated to reduce the surface tension by at least one half. The surface tension of water and the 70.4% glycerol solution thus approached that for the organic liquids and permitted a direct observation of the effect of surface tension without change in viscosity or density. Inspection of Figures 7 and 8 shows that the decrease of surface tension caused no change in film thickness. It was also determined by independent observations that the point of wave inception did not change upon the addition of the surface-active agent.

DISCUSSION OF EXPERIMENTAL RESULTS

The use of radioactive tracers for the measurement of liquid film is a satisfactory technique. Determinations may be made without disturbing the flow of liquid, and a series of values over a wide range of flow rate may be obtained rapidly. The correction for absorption of beta radiation by the liquid and the calibration of the apparatus by observing flow in a region of true viscous flow both provide a procedure for relating radioactivity of the dissolved isotope to the film thickness. The method appears capable of measuring film thickness with a precision of 0.0005 in. or better but the accuracy can only be inferred because an absolute measurement is not possible. The method also has possible application for flow-rate measurement in experimental work involving films.

It is not feasible to calibrate the apparatus by directly relating activity to thickness by use of the planchet technique. Inaccuracies arise from surface-tension effects and reflection of radiation from the planchet walls; also, the geometry of counting a sample in a planchet is different from that in the flow apparatus.

Beta radiation was employed because it is effectively shielded by the walls of the glass and copper tubing, and no other precautions are necessary. An alternate procedure would be to use an isotope emitting gamma radiation, which would be much more penetrating. The absorption would be zero for the thickness of films usually encountered and a linear relation between observed activity and thickness would result; however, this would require additional shielding to reduce the hazard of exposure and to eliminate radiation being recorded from undesired regions of the apparatus. Isotopes emitting beta radiation may be used only where the films being observed are not so thick that absorption effects predominate. A source of gamma radiation could be used where the absorption of beta radiation is large. The choice of isotope is limited to those which are soluble in the liquid under test, or at least to those which can be prepared as a colloidal solution or suspension and preferably to those emitting only one type of radiation.

Statements appearing in the literature(*3, 5, 6*) that liquid films in wave flow exhibit true streamline behavior with respect to thickness have been based on observations for liquids of low viscosity. For liquids more viscous than water the theoretical relations do not hold and the discrepancy can be large. Contrary to a previous report(*8*) the surface tension is not a significant factor in either wave inception or flow pattern when waves are present.

The data permit few conclusions concerning the inception of turbulence. For water and the lighter liquids, no significant departure from viscous behavior is indicated up to Reynolds numbers of 5,000 although greater scattering of the data is observed above $Re = 2{,}000$. Flow rates well up into the turbulent region could not be obtained with the equipment as originally designed.

Equations for Flow Down a Circular Tube. Equations (4) through (8) were derived for flow down a flat plate, and the validity of their use for a circular tube may be assessed by deriving equations for the tube. With reference to Figure 9, for a tube of height h the forces acting on a section of liquid toward the center of the tube at radius r are F_1, the force of gravity acting on the mass of the film; F_2, the force due to viscous shear of the liquid; F_3, the frictional force exerted by the core stream on the liquid surface (flow assumed countercurrent to wall film). Under steady state conditions these forces may be equated as follows:

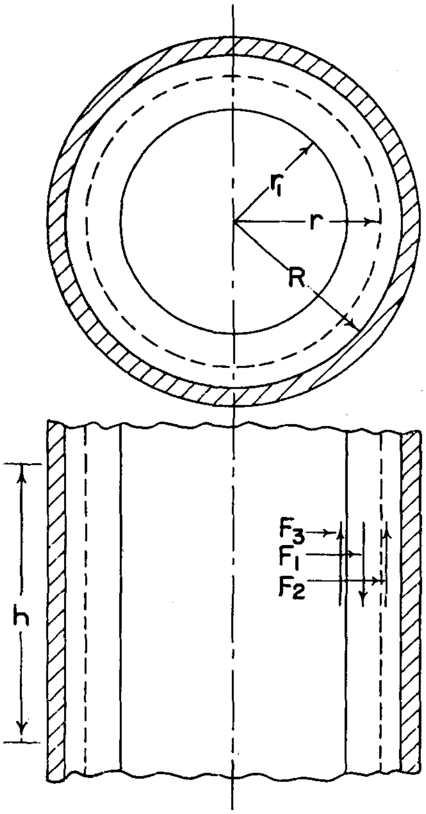

Fig. 9. Sketch Showing Distances and Forces for the Derivation of Equations for Flow of Films Down a Vertical Tube.

$$F_1 = F_2 + F_3 \qquad (9)$$

where

$$F_1 = \pi\,(r^2 - r_1^2)\,h\rho_a g_c \qquad (10)$$

$$F_2 = -\,(2\pi rh)\,\mu\,(dv/dr) \qquad (11)$$

$$F_3 = -\,(\Delta P \pi r_1^2)\,g_c \qquad (12)$$

It has been shown(*10*) for a gas core moving at a moderate flow rate that F_3 is small in magnitude compared with the other forces and may therefore be neglected for this condition.

With this assumption, $F_1 = F_2$ and

$$\frac{dv}{dr} = -\frac{\rho_a\, g_c}{2\mu}\left(r - \frac{r_1^2}{r}\right) \qquad (13)$$

Integrating

$$v = \frac{-\rho_a\, g_c}{2\mu}\left[\frac{r^2}{2} - r_1^2 \ln r\right] + C_1 \qquad (14)$$

The constant of integration may be evaluated by imposing the condition that at

$$r = R, v = 0, \text{or } C_1 =$$

$$\frac{\rho_a\, g_c}{2\mu}\left[\frac{R^2}{2} - r_1^2 \ln R\right] \text{and } v =$$

$$\frac{\rho_a\, g_c}{4\mu}\left[R^2 - r^2 + 2r_1^2 \ln \frac{r}{R}\right] \qquad (15)$$

Equation (15) gives the velocity of a liquid layer at any point r in the film. The liquid surface velocity is given by $v = V_s$ at $r = r_1$, or

$$V_s = \frac{\rho_a\, g_c}{4\mu}\left[R^2 - r_1^2 + 2r_1^2 \ln \frac{r_1}{R}\right] \qquad (16)$$

The mean liquid velocity may be obtained from

$$V = \frac{Q}{\pi\,(R^2 - r_1^2)} \qquad (17$$

where

$$Q = \int_{r_1}^{R} v\,(2\pi r)\,dr \qquad (18)$$

Combining Equations (15), (17), and (18) and integrating give

$$V = \frac{\rho_a\, g_c}{8\mu}\left[R^2 - 3r_1^2 - \frac{4r_1^4}{(R^2 - r_1^2)} \ln \frac{r_1}{R}\right] \qquad (19)$$

Dividing Equation (16) by Equation (18)

$$\frac{V_s}{V} = 2\left[\frac{R^2 - r_1^2 + 2r_1^2 \ln \frac{r_1}{R}}{R_2 - 3r_1^2 - \frac{4r_1^4}{R^2 - r_1^2}\ln\frac{r_1}{R}}\right] \tag{20}$$

The value of this ratio for the limiting conditions of a full and empty pipe may be obtained in each case by applying L'Hopital's rule (*20*). This consists of successively differentiating the numerator and denominator until a determinate value for the ratio is obtained on substitution of limiting conditions. For an empty pipe $r_1 = R$ and $V_s/V = 1.5$, which is the same result as obtained for the flat plate. For a completely full pipe in viscous flow $r_1 = 0$ and $V_s/V = 2$, which is the expected result (V_s in this case is the velocity at the center of the pipe). It is evident that the value of V_s/V will vary continuously between these limits depending on the thickness of the film and the curvature of the pipe wall.

For liquids of medium or low viscosity and for tube radii large in comparison with film thickness, the velocity ratio may be assumed to be 1.5 for most purposes. For instance, water at 25°C. in viscous flow down the wall of a 1.5-in.-diam. tube at a Reynolds number of 2,100 would have a film thickness of 0.020 in. For these conditions the value of the ratio from Equation (20) is 1.505.

Equations (16), (19), and (20) would be required in the case of very viscous liquids, tubes of small diameter, and liquid-liquid systems where film thicknesses are much larger. The less complicated equations for flow down a flat plate would be preferred where applicable.

No quantitative relationship is readily apparent between film thickness and flow characteristics for the region of viscous flow with waves present. The development of a theoretical or empirical description of the flow pattern requires further information at point conditions because of the irregular and constantly changing surface of the film. A qualitative flow pattern and certain quantitative characteristics of wave flow are discussed below.

CRITERION FOR WAVE INCEPTION

In contemplating the behavior of films in wave flow it is apparent that factors other than those for closed conduits must be considered because of the free surface exposed. A means of predicting the conditions under which wave flow occurs is desired.

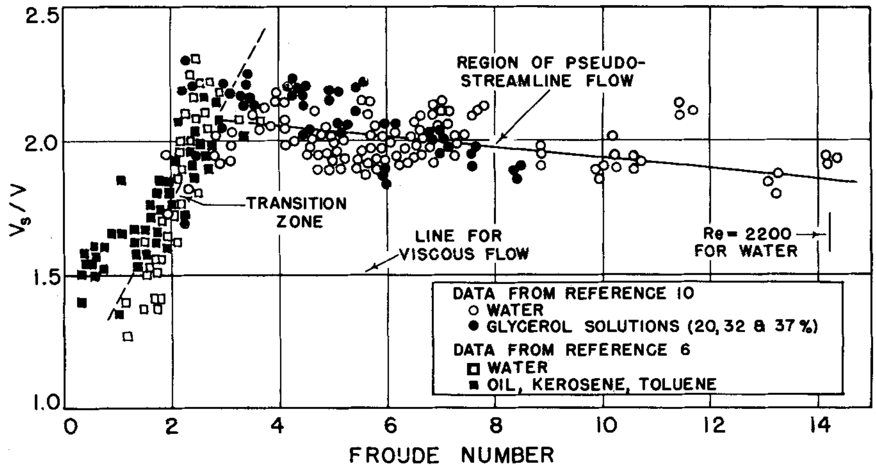

FIG. 10. RATIO OF SURFACE VELOCITY TO AVERAGE VELOCITY AS A FUNCTION OF FROUDE NUMBER FOR THE REGION OF VISCOUS FLOW.

The Weber number may be used to characterize the flow under conditions where surface tension is a factor; however, the results reported show that surface tension is not a variable in either wave inception or film thickness.

The Froude number may be used to characterize the flow when density or gravity is a factor. It may be defined as the ratio of average velocity of the liquid stream to the velocity of propagation of a shallow wave*, or

$$F = V/(g_c m)^{1/2} \tag{21}$$

The velocity of propagation of a wave where the depth of liquid is small is dependent only on the depth and is independent of such factors as viscosity and surface tension(*18*). A Froude number of unity represents a critical condition at which the average liquid velocity is equal to the velocity of wave propagation; it might be expected therefore that this condition would be a critical point for the appearance of waves in film flow. This criterion will first be applied to the available data on surface velocities of liquid layers on vertical tubes.

Surface Velocities. Figure 10 shows the ratio of apparent surface velocity to average velocity plotted vs. Froude number calculated for data from two sources. The method of reference *6* timed the travel of an indicator dye (injected into the film) between two points on the column wall; it was suggested that the region of wave flow be termed pseudostreamline. The method of reference *10* used the pressure drop of a moving air phase in contact with the liquid film to determine surface velocities. The latter method is inapplicable at low liquid flow rates because of the small effect of the film on the air core and the former method is inapplicable at high flow rates because of diffusion and errors in timing. Agreement is evident where the two methods overlap.

Figure 10 shows a region where true viscous behavior is followed, a transition region where the velocity ratio continually increases, and finally a region where the ratio becomes stabilized but shows a small decrease as the limit for viscous flow is approached. The best line for the transition region intersects the line for viscous flow at a value of $F = 1.3$. It was reported(*6*) that more rapid diffusion of the dye was observed in the region where waves were present, which would result in errors of timing. The departure of apparent surface velocities from normal would appear to occur at Froude numbers approximating unity.

Data for Wave Inception. The Froude numbers for wave inception were calculated for four of the systems studied and for data reported by Fallah, Hunter, and Nash(*5*), Grimley(*8*), and Kirkbride(*12*). Results are given in Table 3, where it may be observed that the Froude number for wave

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*The Froude number is sometimes used as the square of this ratio.

formation centers on unity. The data for the two liquid-liquid systems are of particular interest because of the very low apparent density of the wall film. The points given correspond to the first departure from viscous flow and were interpreted by the authors as the beginning of turbulence. The waves were quite pronounced, which may, in fact, lead to a condition of turbulence.

PROBABLE FLOW PATTERN IN WAVE FLOW

The flow pattern assumed by the liquid in the pseudostreamline region must explain a contradictory type of behavior. Except for the more viscous liquids, the film thickness is that for streamline flow; yet the apparent surface velocity is higher than normal. Further, there is the evidence cited for a mixing action in mass and heat transfer processes. Quantitative data have been given(*16*) for the same system in a wetted-wall column where the gas-film resistance was 90 to 100% of the total, compared with a packed tower, where it was only 50%. Visual observations(*6*) by means of a dye technique showed that the color thinned out more rapidly with waves present.

A qualitative explanation for flow with waves is probably somewhat as follows. Waves appear when the Froude number exceeds unity. The wave peak attains such a great height that it becomes a localized point of turbulence. The wave, which moves downward with a velocity greater than that of the laminar film because of the greater thickness, causes a disturbance in the film, which returns to laminar flow when the wave has passed. The film never becomes completely turbulent because the laminar sublayer has appreciable thickness. In Figure 11 the wave crest is likened to a rod rolling over a surface covered with a thin layer of liquid. In support of this picture it must be shown that (1) the wave crest can be in turbulent flow as the criterion is usually applied, (2) the laminar sublayer is relatively thick, and (3) wave volume is small compared with the bulk of the film for liquids of low viscosity but becomes greater as the viscosity increases. Some evidence can be cited.

The data of Kirkbride(*12*) show that at an average Reynolds number of 640 for the film, the Reynolds number based on the wave height would be 8,800, a maximum height for the liquid observed. Reynolds numbers for other flow conditions can be judged by the equation

$$Re = \frac{4 g_c m^3 \, (\rho_a/\mu)^{1/3}}{3} \qquad (22)$$

where the Reynolds number varies as the cube of the thickness. It is usually accepted that the beginning of turbulence in films corresponds to Reynolds numbers of 1,000 to 2,000(*2, 4, 5, 12*).

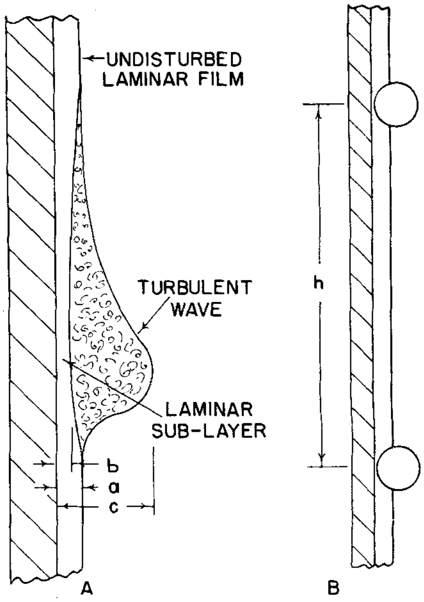

FIG. 11. SKETCH SHOWING PROBABLE PATTERN FOR WAVE FLOW.

TABLE 3.—FROUDE NUMBER FOR WAVE INCEPTION CALCULATED FROM THE DATA OF SEVERAL INVESTIGATORS

Investigator	Properties of wall liquid System*	μ, centipoises	ρ_a, g./cu. cm.	Froude number for wave inception
This paper	Water-air	0.894	0.997	1.12 (avg.)
	2-propanol-air	2.02	0.782	0.92 (avg.)
	62.0% glycerol-air	10.15	1.157	1.23 (avg.)
	70.4% glycerol-air	18.64	1.179	1.06 (avg.)
(*8*)	Glycerol *A*-air	26.4	1.203	0.82
	Glycerol *B*-air	12.1	1.184	0.87
	38.7% Ethanol-air	2.84	0.942	0.87
	92% Ethanol-air	1.52	0.817	0.90
	62% Ethanol-air	2.60	0.891	1.01
	Carbontetrachloride-air	2.62	1.6	0.74
	Benzene-air	0.70	0.874	1.17
	Water-air	1.12	1.0	1.44
(*12*)	Oil 1-air	17.6	0.88	0.70
	Oil 3-Air	1.95	0.82	1.08
(*5*)	Oil-water	1.31	0.128	0.83
	Kerosene-water	1.31	0.183	1.29

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*Wall liquid given first

Even though the bulk of a stream may be in turbulent flow, the portion near a solid boundary remains in laminar motion(*14*). Discrepancies have been shown to exist in the generalized velocity-distance relationships as first proposed(*15*), but these may be used to illustrate the nature of the laminar sublayer in a semiqualitative manner. As pointed out by Colburn(*2*) there exists a value of the Reynolds number below which the flow will be all viscous. For higher flow rates the laminar sublayer has a minimum thickness corresponding to this limiting condition, and the remaining portion of the film need not necessarily be all viscous. In the presence of an external force acting on the film (in this case the weight of the turbulent wave) the limiting Reynolds number was calculated to be 270. Reference to Figure 11A and an example will make this clear.

For water flowing as a film down a vertical tube at an average Reynolds number of 640 (room temperature) the undisturbed thickness *a* would be 0.012 in., the sublayer *b* would be 0.009 in., and the height of the wave crest *c* from Kirkbride's data would be 0.029 in. These figures, though only approximate, serve to show that the disturbance caused by the passing wave is smaller than might be expected.

The laminar sublayer is probably not strictly viscous in all respects. Lin(*13*) has pointed out that disturbances in the transition zone (between turbulent and laminar layers) may propagate sidewise into the laminar layer and that a small mixing action may result.

No accurate data are available for wave volume but a simple assumption permits an estimate of the relative volume of wave and film. If the wave volume is assumed to be equal to that of a cylinder of diameter equal to wave height, and if wave spacings of 1 1/4 in. for water and 1 1/2 in. for the more viscous liquids at low flow rates are assumed, the ratio of film volume to wave volume for water would be 62, but only about 5 for the more viscous glycerol solutions. As the flow rate increases, the ratio decreases for either case but would be more pronounced with the viscous liquids, as observed experimentally.

A quantitative explanation of the flow behavior requires data on wave velocity, profile, and spacing, which may be obtained by a recently developed technique(*4*). The complexity of the problem is indicated by the highly irregular and changing character of the waves (references *4* and *8* show some excellent photographs) and the ultimate solution may require a statistical treatment. It is possible that the flow pattern of falling films in the turbulent region may also involve wave flow and that basic information for the viscous region can be applied in describing flow in the turbulent zone.

APPLICATIONS

The increase of surface velocity for the wave-flow region arises because of the increased velocity of the waves. The weight of the wave exerts a force on the boundary of the laminar sublayer and causes the normal parabolic velocity gradient [Equation (5)] to become more nearly linear. Thus the local velocity of fluid in the laminar sublayer is greater than normal and the velocity of the wave must be determined to some extent by the velocity at the boundary. The ratio of surface velocity to average film velocity is 1.5 for the parabolic gradient but 2.0 for a linear gradient and Figure 10 shows the ratio for the pseudostreamline region to be about 2.0. However, the assumption of a linear gradient for the bulk of the film is inconsistent with the observed thicknesses, and the surface velocities observed are only apparent and represent an average effect of wave flow and surface roughness. It is also possible that the interfacial area exposed with wave flow is significantly greater than for undisturbed laminar flow (*4*).

A useful form for the Froude number may be obtained by substituting the value of V from Equation (7) in Equation (21) and eliminating m by Equation (4):

$$F = \left[\frac{Q\ \rho_a}{3\mu} \right]^{1/2} \qquad (23)$$

With the condition imposed that at wave inception $F = 1$, the flow rate at which waves appear depends only on the kinematic viscosity, or

$Q\text{(for wave inception)} = 3(\mu/\rho_a)$

Thus waves will form at low rates for low viscosity liquids, but higher rates are necessary to establish wave flow as the viscosity increases. This behavior was observed for the liquids employed. For liquid-liquid systems, where the ratio of (μ/ρ_a) is large, waves will appear in the wall liquid at a flow rate greater than that for a corresponding liquid-gas system. Nevertheless, the point of wave inception for liquid-liquid systems probably lies well within the usual operating range.

The Froude number is used as a criterion for vortex formation in mixing problems(*19*) and as a means of predicting particulate or aggregative fluidization in fluid-solid systems(*23*). It thus becomes an additional parameter in the fluid mechanics of processes important to the field of chemical engineering.

In correlating mass transfer coefficients with fluid properties for two phase systems, the effect of each phase on the fluid mechanics of the other must be considered. For instance, in liquid-gas systems the Reynolds number of the gas phase must be that relative to the liquid surface, and the velocity of the gas stream calculated from the mass rate of flow must be increased (if countercurrent) by an amount equal to the liquid surface velocity. This correction can amount to 25% or more for rectification(*9*) and for air-water(*10*). Figure 10 may be used to determine the apparent surface velocities of low-viscosity liquids in the pseudostreamline region and as a first approximation for more viscous liquids. It is possible that the behavior shown in Figure 10 is not typical for high-viscosity liquids or liquid-liquid systems or for the case where the force, F_3, at the interface is not negligible.

The behavior pattern of liquid films where the drag force at the interface is not negligible would depend on the magnitude and direction of this force. For countercurrent flow (*see* Figure 9) the average liquid rate would be decreased; for cocurrent flow the drag force and gravity would act in the same direction, thus speeding the flow of the wall liquid. The point of wave formation and the velocity gradient in the film would be influenced. The presence of a finite force, no matter how small, can easily be demonstrated by blowing smoke across the top of a column of liquid which exposes a free surface. A portion of the smoke is rapidly transported to the bottom of the column. Data have been presented(*10*) which show that the drag force can be neglected for moderate gas velocities. The magnitude of the force can be calculated from the pressure drop if this is known and if the geometry of the flow channel is simple. Colburn(*2*) has described the unusual behavior of the liquid film for a system where the vapor is moving at very high velocities.

In translating the behavior of films in the pseudostreamline region to the flow of liquids in packed columns it should be pointed out that the free fall necessary to establish wave flow may not occur in this type of equipment because of the discontinuous flow path. It was shown earlier that waves did not appear for some distance below the inlet for a wetted-wall column. The absence of wave flow in a packed column, or the extent to which it does occur, should be verified. It was observed(*9*) for rectification in a wetted-wall column that discontinuities in the height of a transfer unit occurred in the upper part of the column, indicating that some liquid-film resistance was present where wave flow was not established. Peck and Wagner(*16*) showed that liquid-film resistances were small for a wetted-wall column but comprised half the total resistance for packed columns.

Industrial process equipment could be designed to take advantage of the low liquid side resistances of falling films in the pseudostreamline and turbulent regions. The use of a bank of vertical wetted-wall tubes was reported to be unsatisfactory because of inadequate liquid distribution, but it is conceivable that a series of vertical plates, each with an independent supply of liquid, would provide a large surface area and would permit large quantities of liquid and gas to be brought into contact. Such a system would be particularly attractive for gas-absorption systems because of the low pressure loss in the gas phase. The use of a falling film inside a vertical tube with an external supply of heat has been used for a stripping operation. A new technique in gas-separation processes, "sweep diffusion" (*1*), provides a recent application of the use of liquid film for component separation.

SUMMARY AND CONCLUSIONS

Equipment and a method are described for the measurement of thickness of liquid films by the use of a radioactive tracer dissolved in the liquid. The method has a precision of 0.0005 in. and could be applied for the further study of liquid films in gas-liquid or liquid-liquid systems. Six liquids with viscosities ranging from 0.5 to 20 centipoises were observed in flow down the inner wall of a vertical tube.

Wave motion appears at low flow rates well within the viscous region, which results in departure from true viscous flow. The waves are shown to appear when the Froude number exceeds unity. Liquids having a viscosity less than that of water exhibit the film thicknesses expected for true viscous flow even though waves are present. Liquids with a viscosity greater than that of water show film thicknesses less than for viscous flow. The departure from normal behavior increases with increasing viscosity. Surface tension is not a factor in either wave inception or flow behavior with waves present.

Equations for the flow of viscous films inside tubes (without waves) are derived and would be required for small-diameter tubes or very viscous liquids. In many situations, particularly those involving low-viscosity liquids, the less complex equations for flow down a flat plate may be used.

A qualitative theory is proposed which accounts for the presence of a pronounced mixing action even though the basic flow is essentially streamline. The waves attain a condition at low average flow rates where turbulence can begin locally. The bulk of the film and the sublayer beneath the wave remain in streamline flow. The turbulent wave moves downward at a velocity greater than that of the film and creates a disturbance and mixing action in passing. The ratio of wave volume to film volume is small except in the case of liquids more viscous than water.

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NOTATION

A, A' = activity of a sample of liquid as analyzed and as corrected to a given date, counts/sec.

A_o, A_c = activity of liquid film on vertical-tube apparatus as observed and as corrected for absorption and sample activity, counts/sec.

C = constant relating corrected activity to film thickness, ft./(counts/sec.)

F = Froude number, dimensionless, equal to $V/(g_c m)^{\frac{1}{2}}$ for a liquid film

F_1, F_2, F_3 = forces acting on an isolated portion of film, lb. force

g_c = dimensional constant, 4.170×10^8, (lb. mass) (ft.)/(lb. force) (hr.^{2})

h = height of column section, also spacing between successive waves, ft.

m = film thickness, ft.

ΔP = pressure drop over height h of column, lb. force/sq.ft.

P_a, P_c = absorption and concentration factors, defined in the text

Q = volumetric rate of liquid flow per unit of wetted periphery, cu.ft./(ft.) (hr.)

r, r_1 = radius from center of tube to point within film and to the inner surface of the film, ft.

R = radius of tube and outer boundary of film, ft.

Re = Reynolds number $= 4Q\rho_a/\mu$ for a liquid film

t = time of radioactive decay, days

v, V, V_s = point, average, and surface velocity, ft./hr.

x = distance from wall to point within film, ft.

λ = constant for radioactive decay, days^{-1}

μ = liquid viscosity, lb. mass/(ft.) (hr.) unless given otherwise

ρ_a = apparent film density, equal to liquid density minus fluid density of core (the latter is negligible for a gas core), lb. mass/cu.ft. unless given otherwise

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