TURBULENCE IN FALLING LIQUID FILMS

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On the basis of fluid dynamic and heat transfer studies on falling-film towers by various investigators, it has been commonly accepted by most workers that the liquid flow is essentially streamline in nature for liquid-film Reynolds numbers under 1,800 to 2,000; consequently it would be expected that the rate of physical gas absorption in such liquid films could be predicted directly from a knowledge of molecular diffusion rates.

Measurements of the absorption of pure gases in falling liquid films at low Reynolds numbers substantiated the findings of other investigators that the mass transfer rates were manyfold greater than could have been predicted if molecular diffusion were the only transfer process. Increased interfacial area due to rippling of the liquid films could not account for the large increase in mass transfer rates found, and experiments with the addition of a dye stream to the liquid at the free interface indicated turbulence.

Dissolution rates of slightly soluble solids coated on the tube wall to liquid films were measured and showed that the liquid film was not in laminar flow even for Reynolds numbers as low as 300.

An explanation is proposed which resolves these apparently conflicting results between momentum and heat and mass transfer, based on the fact that mass transfer measurements provide a more sensitive test for the presence of turbulence than do momentum or heat transfer measurements.

Existing theory for the design of gas-liquid reactors is not adequate at the present time owing to lack of knowledge as to what is taking place in the liquid phase near the phase boundary. This work was undertaken to study the physical system, without reaction, since an understanding of the mechanism of liquid-phase mass transfer is essential for the development and application of any fundamental theory for the combined diffusion-reaction case. A falling-film tower was selected for preliminary work

primarily because of its relatively constant and known interfacial area. In addition, certain apparent anomalies exist between mass, heat, and momentum transfer data for falling-film towers, and it was believed that an explanation which could reconcile these differences would also shed light on the liquid-phase mass transfer process.

LIQUID-PHASE MASS TRANSFER DATA

A number of investigators have studied mass transfer in falling liquid films and have in general found that the measured absorption rates are considerably greater than would have been predicted by theory. For example, Johnstone and Pigford (14) reported values for the heights of liquid-film transfer units, H_L , for distillation in a wetted-wall column one half to one quarter those predicted by theory for liquid-film Reynolds numbers ranging from 100 to 500. In a paper on distillation of ethanolwater in a wetted-wall column, Surowiec and Furnas (25) reported

heights of liquid-film transfer units about 1/14 of those predicted from heat transfer equations in the Reynolds number range of 125 to 340. Data from the work of Hodson(11) and Hurlburt(12) on oxygen and carbon dioxide desorption in a 1.05- by 18.5-in. tower show desorption rates severalfold greater than theory in the laminar flow range. Runs made in this investigation on the absorption of carbon dioxide in water in roughly 3- and 6-ft. towers also indicated mass transfer rates 1 1/2- to 3-fold as great as the rate predicted by theory based on molecular diffusion below liquid-film Reynolds numbers of 1,800. The apparatus of all investigators differ somewhat in the types of calming sections used for liquid and gas introduction, but this detail does not appear to be significant in the relatively long towers used. One thing common to all the liquid-film work appears to be the presence of ripples in the liquid film, and this fact has been used by previous investigators to explain the higher than expected mass transfer results.

MOMENTUM-TRANSFER DATA

The equations for viscous flow in a falling-film tower with no drag at the free interface were first set up by Nusselt (22). The theory shows that the velocity distribution is parabolic, with the maximum velocity at the free interface equal to 1.5 times the average velocity. Friedman and Miller(8) have shown that the surface velocity is considerably greater than that predicted by theory at Reynolds numbers above 25, about the point they found rippling to begin. Experiments by Grimley (10), in which high-speed photographs of illuminated particles within the liquid film were taken to determine the velocity profile at different points, indicated that the liquidvelocity profile goes through a maximum about 1/10 of the film thickness from the gas-liquid interface. This resembles the velocity profile commonly encountered in open channel flow. Near the wall, however, the velocity profile measured by Grimley does not depart appreciably from that of a parabolic gradient. Although these experiments point out discrepancies between the theory and actual conditions, nevertheless a number of investigators have shown (5, 7, 22)that the liquid-film holdup checks the holdup predicted by the theoretical equations up to Reynolds numbers of 1,800 to 2,000. This

range of Reynolds numbers has, therefore, been commonly used as the criterion for the presence or absence of turbulence; i.e., above this range the equations cease to hold because of the presence of turbulence.

HEAT TRANSFER DATA

In addition to work on momentum transfer, a number of workers have carried out experiments on the condensation of pure vapors on the outside surfaces of vertical tubes (16, 19, 21). In general, below a Reynolds number of 2,000, the data for heat transfer rates fall 25 to 50% higher than values predicted by the viscous-flow equations based on a parabolic velocity distribution.

Data have also been reported (1) for the heating of fluids in streamline flow in falling-film towers, where heat is transferred from a wall at constant temperature to the fluid, care being taken to avoid appreciable vaporization. For the streamline flow region (based on the criterion 4 $\Gamma/\mu \leq 1,800$) the data fall some 20 to 30% higher than predicted by theory.

DEVELOPMENT OF THEORY

The previous paragraphs present the experimental facts on hand at the start of this work. The question to be answered was why mass transfer rates were so much greater than those predicted by theory in the streamline region, while holdup measurements checked laminarflow theory closely and heat transfer results, while slightly higher than theory, were nevertheless in fair agreement. The data for momentum and heat transfer are consistent with the postulation of laminar-flow conditions, and the data for the mass transfer case indicate the presence of turbulence in the liquid film.

The explanation used by previous investigators for the high rate of mass transfer in the streamline flow region has been associated with rippling of the liquid film, which could increase mass transfer results by increasing the interfacial area or by creating turbulence* in the liquid film. The maximum increase in surface caused by the presence of ripples as judged by visual observation and photographs is probably less than 50%, and this would not be adequate to explain the increased rates observed in the mass transfer experiments. The work of Friedman and Miller(8) indicated turbulence in the liquid at the free interface, and similar experiments were performed by the authors. A dye stream introduced on the surface of a water film was observed to spread out in the form of a cone downstream from the point of introduction. At no time was a laminar dye thread observed in the rippling film although the Reynolds number was reduced to as low as 200. Thus the evidence at hand indicated turbulence at the free interface, which would be expected to be present to some degree within the main liquid film. The question raised is whether or not turbulence can markedly increase mass transfer rates while not affecting to any major degree either momentum or heat transfer rates.

Equations of definition can be written for molecular transport of momentum, heat, and mass as

Momentum

$$\frac{1}{\rho} \frac{dP}{dL} = \frac{1}{yg_c} \frac{\mu}{\rho} \frac{dV}{dy}$$

Heat

$$\frac{1}{a} \frac{dT}{d\theta} = \frac{k}{\rho c_p} \frac{dT}{dy}$$

Mass

$$\frac{1}{a} \frac{dc}{d\theta} = D_L \frac{dc}{dy}$$

It will be noted that μ/ρ , $k/\rho c_p$, and D_L all have identical units and can be considered as rates per unit driving potential; i.e., per unit force, temperature, and concentration. For water at 31°C. these diffusivities have the following values:

Momentum transfer (kinematic viscosity) = $3,020 \times 10^{-5}$ sq.ft./hr.

Heat transfer (thermal diffusivity) = 569 × 10⁻³ sq.ft./hr. Mass transfer (molecular dif-

fusivity) $D_L \text{CO}_2$ in water = 9.3 \times 10-5 sq.ft./hr.

 D_L benzoic acid — water = 5.2 \times 10-5 sq.ft./hr.

From the foregoing tabulations it can be seen that for water films the momentum diffusivity is about five times as large as the thermal diffusivity, which is in turn about sixty times as large as the molecular diffusivity. These values change slightly with temperature, but the ratios between them remain essentially the same. These significant differences in transfer rates

^{*}The criterion for turbulence used throughout this paper is based upon whether or not experimental results check mass transfer theory based solely on molecular diffusion.

per unit driving force between momentum, heat, and mass transfer for liquid films are not surprising, since mass transfer is the transport of a specific molecular or ion, while heat and momentum may be transferred not only by physical transport, but also by collision. From this, it is apparent that if turbulence were present to any degree, it would have a greater effect upon the mass transfer rate than upon the rates of heat or momentum transfer; i.e., the slowest process would show a measurable effect first. With this assumption it then would be possible, within a given range of Reynolds numbers, to have equations based on laminar flow hold reasonably well for momentum and heat transfer and not correlate mass transfer data at all.

It is of interest to examine the diffusivities for gas films. For air at 0°C. and atmospheric pressure, the diffusivities have the following values:

Momentum transfer (kinematic viscosity) = 0.51 sq.ft./hr.

Heat transfer (thermal diffusivity) = 0.69 sq.ft./hr.

Mass transfer (molecular diffusivity) D_V CO₂ in air = 0.53 sq. ft./hr. D_V water in air = 0.85 sq. ft./hr.

It is immediately noted that the large differences in diffusivities for momentum, heat, and mass transfer disappear for gas films. It would thus be expected that correlations (3, 9, 24) between heat and mass transfer for gas systems in turbulent flow would be valid through the dip region, while similar correlations for liquid systems would not be expected to hold in this manner.

The foregoing explanation, although forming a logical explanation for the observed abnormalities in momentum, heat, and mass transfer rates in liquid films, needs proof. An experimental program was, therefore, set up to furnish data to prove or disprove the proposed explanation.

EXPERIMENTAL WORK

As it was desired to obtain mass transfer data under identical physical conditions employed by previous workers (1) in obtaining heat transfer data, the rate of solution of various materials from a solid wall to a falling liquid film was measured. In addition to enabling one to establish whether turbulence existed below Reynolds numbers of 1,800 to 2,000 in falling liquid films near the tube wall, these measurements would also give a quantitative picture of the

diffusivity across the film from the wall side outward. Together with absorption measurements previously referred to, this would yield a rather clear picture of the extent of turbulence existing across the entire film.

The first three materials to be used as coatings were benzoic acid, adipic acid, and succinic acid. The tubes were coated internally by melting the crystalline acids and pouring molten liquid into the tubes via a funnel while rotating the tubes. In this simple manner a uniform and smooth coating was obtained. It was found that fine crystals and, therefore, smoother surfaces were obtained if cold water was circulated through a jacket surrounding the tubes so that rapid crystallization took place.

The falling-film tubes used in the experiments were jacketed glass with the inside diameter as measured with the solid coating in place varying from 0.77 to 1.13 in. Column lengths varied from 30 to 74.5 in. The liquid was introduced into a reservoir at the top of the columns and flowed over the top. Several layers of stainless steel screen held in place around the top of the column caused a slight head of liquid to build up in the reservoir above the tube inlet, and in this manner uniform liquid flow around the periphery of the tube was obtained. Although no definite calming section at the liquid entrance was provided, the solid coatings all started some 3 to 4 in. from the top of the column.

Distilled water was used in all the runs and this was heated to the operating temperature by means of a coil immersed in a constant-temperature water bath. In all runs the temperature change of the water between inlet and outlet was no more than ±0.2°C. Analysis of the effluent stream for benzoic acid, adipic acid, and succinic acid, was made by titrating with standard sodium hydroxide using phenolphthalein as indicator. The percentage saturation of the effluent as reported in the tables of data is believed to be accurate within 10%, the precision as measured by check samples being within 1%. The solubilities of the acids in the water used were measured and these figures were used in all calculations. A comparison of measured solubilities with values from the literature, where available, checked closely. Molecular diffusivities used for comparison with the measured apparent diffusivities were taken from the literature (13,

It was found that water would not completely wet the benzoic acid surface below Reynolds numbers of about 3,400. A wetting agent, the sodium salt of an aliphatic sulfonate, was then added to the water (0.02 and 0.05% by weight) and runs were made down to a Reynolds number of about 950. Below this Reynolds number, incomplete wetting was again encountered and it was found that adding additional wetting agent did

no further good.

When results of the runs at Reynolds numbers from 2,200 to 3,000 made with the wetting-agent-treated water were extrapolated to Reynolds numbers of 3,400 to 3,800, they were found to check closely results obtained in the latter Reynolds number range using pure water. Thus the addition of the small amount of wetting agent appeared to have a negligible effect upon the dissolution rate. All adipic and succinic acid runs were made with 0.05% by weight of wetting agent in water.

Since it was anticipated that there might be some question as to how smooth the coatings of benzoic, adipic, and succinic acids were, two other systems were run; i.e., paraffin waxbutanol and stearic acid-ethanol. Although both of these latter solids are crystalline, the crystals are small and an extremely smooth surface can be produced by the coating method used. In addition, no trouble was encountered with wetting of the solid surfaces by the liquids and no wetting agent was therefore employed. Analysis of the effluent in the paraffin wax -butanol system was made gravimetrically by evaporating the butanol. With the stearic acid-ethanol system the stearic acid in the effluent was determined volumetrically by direct titration with sodium methylate using phenolphthalein as indicator. Values of the molecular diffusivity were obtained from the literature (13) or estimated by the method of Wilke (27).

TREATMENT OF DATA

The differential equation set up by Nusselt (22) for the flow of heat from a wall at constant temperature to a falling liquid film is

$$-k \frac{\partial^2 T}{\partial u^2} c_p V \rho \frac{\partial T}{\partial L} = 0 \quad (1)$$

Since V varies with y, Nusselt substituted the parabolic velocity distribution, shown to be accurate from holdup data, to obtain

$$k \frac{\partial^2 T}{\partial y^2} = \frac{C_p \rho^2}{\mu} \left(yB - \frac{y^2}{2} \right) \frac{\partial T}{\partial L} \quad (2)$$

Nusselt solved the foregoing equation by a difference method and tabulated his results as ϕ vs. S where

$$S = \frac{k\mu L}{C_{ro}^{2}B_{q}^{4}}$$
 and $\phi = \frac{T_{2} - T_{1}}{T_{o} - T_{1}}$

Nusselt also shows his results as a plot of ϕ vs. S, which is reproduced in Figure 1. The differential equation for mass transfer applica-

ble to the dissolution runs may be obtained by substituting concentration for temperature and D_L , the diffusivity, for $k/c_p \varrho$.

The differential equation for absorption of a gas in a falling water film with a parabolic velocity distribution is

$$\left[1 - \left(\frac{s}{B}\right)^2\right] \frac{\partial C}{\partial L} = \frac{D_L}{Vm} \frac{\partial^2 C}{\partial s^2}$$
 (3)

Johnstone and Pigford (14) give the following solutions of the Equation (3):

$$\frac{C_2 - C_i}{C_1 - C_i} = 0.786 e^{-5.12\eta} +$$

$$0.100 e^{-39.3 \eta} + 0.036 e^{-105.6 \eta} + \dots$$

(4)

where

$$\eta = \frac{D_L L}{B^2 V m}$$

for low values of η the series may be expressed as

$$\frac{C_2 - C_i}{C_1 - C_i} = 1 - \frac{3}{\sqrt{\pi}} \sqrt{\eta}$$
 (5)

The foregoing solutions were used together with the experimental data to calculate the apparent liquid-film diffusivities for the carbon dioxide absorption and solid dissolution runs. The equations were used throughout the Reynolds range studied, since the data of Grimley (10) indicate a maximum deviation of about 30% between measured and calculated velocities at various points within the film.

RESULTS

Experimental results for the solid dissolution runs and the CO_2 absorption runs referred to under Mass Transfer Data are shown plotted in Figure 2 as the apparent liquid diffusivity vs. Reynolds number. As can be seen, the apparent diffusivities for the CO_2 absorption and solid dissolution runs are considerably greater than the molecular diffusivities, which vary from about 0.5×10^{-5} sq.ft./hr. to 10×10^{-5} sq.ft./hr. This is so down to Reynolds numbers as low as 300 to 400.

It will be noted that the apparent eddy diffusivities have been plotted against Reynolds number in Figure 2. The usual equation of definition for turbulent mass transfer is

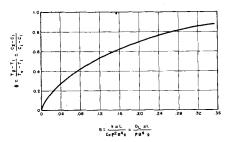


Fig. 1. Plot of ϕ vs. S, from Nusselt (22) for Transfer of Heat or Mass from a Wall to a Falling Liquid

$$\frac{1}{a}\frac{dc}{d\theta} = (D_L + E_D)\frac{dc}{dy}$$

The equation states that the apparent diffusivity is the arithmetic sum of the molecular and eddy diffusivities. It is readily seen that if this is so the data indicate that the true eddy diffusivity is a function of the material being trans-

ferred, which does not appear at all reasonable. The authors prefer to believe that while the preceding equation holds with negligible error where D_L is much larger than E_D or vice versa, it is not valid where D_L and E_D are of the same order of magnitude.

It is apparent from the results that one is dealing with turbulent flow and that as far as mass transfer is concerned turbulence appears to persist throughout the flowing layer. The conclusion which may be drawn is that the assumption of turbulence throughout the liquid layer leads to negligible error in the calculation of mass transfer rates both for the absorption process and the solid solution process. This conclusion is not only in contrast to the usual assumption of turbulent, buffer, and completely laminar zones for fully developed turbulent flow (2, 18, 23, 26), but also differs somewhat from the assumption made for heat transfer

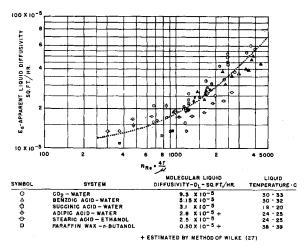


FIG. 2. VARIATION OF APPARENT LIQUID DIFFUSIVITY WITH REYNOLDS NUMBER.

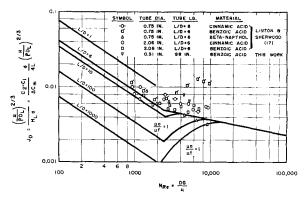


FIG. 3. LIQUID-PHASE MASS TRANSFER IN FULL TUBES.

by Murphree (20), who assumed that the thermal eddy diffusivity varied as the cube of the distance from the wall at small distances from the wall.

A comparison of the results of the mass transfer runs with those of the published heat transfer runs seems to prove that mass transfer rates are affected considerably more than heat transfer rates by small amounts of turbulence. Further, since holdup data for fallingfilm towers are correlated well up to a Reynolds number of 2,000 by use of the theoretical laminar flow equation, it is apparent that momentum transfer is not affected by turbulence to so great an extent as either mass or heat transfer. It is postulated that the presence of turbulence is not necessarily proved by measurements of any singular effect and that the transition of flow from laminar to turbulent is noted at decreasing Reynolds numbers when measurements of momentum, heat, and mass transfer rates are respectively used as criteria. It is of interest to recall that the experiments of Reynolds, using the diffusion of a dye as a criterion for turbulent flow, are visual measurements of mass transfer rates.

SURFACE TENSION EFFECTS

There is one more variable not mentioned up to this point which markedly affects film appearance and mass transfer rates in fallingliquid films. This variable is surface tension. Experiments were conducted in which a stream of dye was introduced into a water film through a 0.013-in, hole in the wall of a 1 1/2 in. I.D. Lucite tube, about 3 ft. long. The water film was found to ripple at all Reynolds numbers and the dye thread was dissipated within 12 to 14 in. from the point of introduction. Some of the wetting agent previously referred to was then added to the water. Not only did the dye thread become continuous throughout the entire tube length when the wetting agent was added, but also the physical appearance of the film changed, the ripples disappeared, and the film became physically laminar even up to Reynolds numbers of about 3,000. If the tube was tapped with the hand, ripples immediately appeared but in a few seconds were completely damped and the dye thread again became continuous. These observations indicate that if ripples are eliminated, mass transfer rates into the liquid should check theory. This

has been shown to be the case in the work of Emmert and Pigford (6).

The possibility exists that the use of the wetting agent in most of the organic acid—water systems may have affected the dissolution results to some extent. Ripples were present in all the dissolution experiments. Since rippling of the water film was not visually affected in these systems and since the results check reasonably well the data for the systems where no wetting agent was used, it is believed that any correction necessitated by the use of the wetting agent is small.

MASS TRANSFER FROM A COATED WALL TO A LIQUID IN FULL

In full pipes the Reynolds number criterion is well defined, and up to the breakpoint, equations based on laminar flow correlate mass transfer data well(17). Linton and Sherwood (17) also have shown that mass transfer rates for liquid systems follow reasonably well the Chilton and Colburn correlation(3) both in the transition and turbulent flow ranges. (The exception to this would appear to be the benzoic acid-water system. which gave higher mass transfer rates than the other two systems studied by Linton and Sherwood; however, these authors attributed the higher rates to cracks which developed in the cast pieces which formed the tube wall.) Since the Chilton and Colburn correlation was developed from heat and mass transfer measurements with gases. the reasonable check of theory afforded by the Linton and Sherwood data would appear to refute the reasoning used previously which proposed that differences between heat and mass transfer for liquid systems should be evident,

Once the method of coating benzoic acid on the interior walls of tubes was developed, two runs were made on the transfer rates of benzoic acid to distilled water in a full tube 0.310 in. I.D. by 99 in. long. The Reynolds numbers at which the runs were made were intentionally selected just above the critical point so that turbulence, although not fully developed, would be present. The mass transfer rates predicted by the Chilton and Colburn correlation (3 and 4) in the dip region were originally taken from heat transfer measurements, to which a correction was added for free convection due to density and viscosity difference. In the light of the falling-film work, it would be expected that for full pipes the actual mass transfer rate would be higher than that predicted from heat transfer measurements at Reynolds numbers where turbulence had just started. The measured mass transfer rates for the two runs, calculated as j factors, were found to be 2 to 2 1/2 fold as great as those predicted by the Chilton and Colburn correlation (see Figure 3).

A possible explanation of the differences between the Linton and Sherwood results and the two runs made in this work is based on an analogy from heat transfer work with gases. When heat is transferred through a laminar boundary layer where there is a stepwise wall - temperature variation, "starting-length correction factor" is introduced to correlate the data. In effect the heat transfer coefficient at a point along the heated surface is found to be lower when the heated section follows a surface which is not heated than when it does not. This decrease in heat transfer is a function of the ratio of the distance along the heated surface to the total distance along the surface and would be more pronounced for short heated sections. While calculations for a short heated length in a long tube (similar to mass transfer apparatus used by Linton and Sherwood) show that this type of correction would be small for gases, it is nevertheless in the right direction. If, as proposed above, the effect is more pronounced for mass transfer in liquid systems, then the differences between the mass transfer data may be reconciled.

SUMMARY

An explanation for the apparent conflict in the data for momentum, heat, and mass transfer in fallingliquid films below a liquid-film Reynolds number of 2,000 is proposed. The results obtained indicate that apparently a slight amount \mathbf{of} turbulence throughout what has heretofore been thought to be a laminar film. While the increase in transfer rates due to turbulence in the film is slight for heat and momentum transfer, this increase is important for mass transfer. The apparent diffusivities herein reported for falling films can be used directly in the equations for the diffusionreaction case, which have been predicated on the basis of a constant diffusivity throughout the liquid. It is suggested that this technique be used as a means for studying turbulence close to walls and phase boundaries.

Although this work is confined for the most part to falling-liquid films, the effect of turbulence upon liquid-phase mass transfer rates would be expected to be qualitatively the same for liquid-phase diffusion in other types of equipment. Thus it is indicated that the assumption of a constant diffusivity from the phase boundary to the bulk of the liquid may be a much closer approximation of the true facts than the sharp demarcation between molecular and eddy

diffusivities commonly used. Although this concept does not add measurably to present methods for handling liquid-phase physical diffusional problems, the simplification which may be possible would greatly aid in the solution of the diffusion and simultaneous reaction problem.

TABLE 1—PHYSICAL ABSORPTION RUNS, 100% CARBON DIOXIDE—WATER*

| | | Tower: 1.0 in. I. | $D. \times 76.5$ in. long | 67 6-4 | Apparent |
|------------------|------------------------------|------------------------------|------------------------------|------------------------------|---------------------------------|
| Run | $4\Gamma/\mu$ | Liquid Temp. | Gas Temp. | % Saturation of exit liquid | diffusivity, sq. ft./hr.×10⁵ |
| 1 2 3 4 | 2500 1270 2500 3750 | 30.4 33.0 31.3 30.2 | 30.0 31.1 31.2 31.0 | 44.0 50.5 42.2 37.4 | 48.4 25.9 44.5 60.4 |
| | | Tower: 0.852 in, | I. D. ×36 in. long | | |
| 5 6 7 8 | 380 690 690 1270 | 32.0 31.2 31.5 31.4 | 30.4 30.5 30.5 30.6 | 56.8 41.5 48.1 30.6 | 13.2 15.5 20.9 19.4 |

^{*}Complete table may be obtained as document 4560 from the American Documentation Institute Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington 25, D. C., for \$1.25 for photoprints or microfilm.

TABLE 2.—MASS TRANSFER OF BENZOIC ACID FROM A WALL TO A FALLING WATER FILM*

| Run | $4\Gamma/\mu$ | Avg. water temp. | Normality exit liquid, °C. | Normality saturated solution | % Saturation of exit liquid | Molecular diffusivity (lit. value), sq. ft./hr. | Apparent diffusivity (Nusselt Eq.) sq. ft./hr.×10 ⁵ | Weight % wetting agent added |
|-----|---------------|------------------------|----------------------------------|------------------------------------|-----------------------------------|---|---|---------------------------------------|
| | | | Tower: 74.5 in | . coated length | 0.91 in. I. D. | | | |
| 1 | 2410 | 30.5 | 0.00475 | 0.0261 | 18.2 | 5.15×10^{5} | 30.0 | 0.05 |
| 2 | 1670 | 31.0 | 0.0055 | 0.0261 | 21.1 | 5.15×10^{5} | 23.1 | 0.05 |
| 3 | 1000 | 31.0 | 0.00765 | 0.0261 | 29.3 | 5.15×10^{5} | 19.1 | 0.05 |
| 4 | 965 | 31.0 | 0.0079 | 0.0261 | 30.3 | 5.15×10^{5} | 18.6 | 0.05 |

^{*}See footnote to Table 1.

TABLE 3.—MASS TRANSFER OF BENZOIC ACID FROM WALL TO WATER, FULL PIPES

| Run | DG/u | Avg. water temp., °C. | Normality exit water | Normality saturated water | % Approach | L/D | $N_{SCH},\ \mu/ ho D_L$ | j_D |
|-----|------|-----------------------------|----------------------------|---------------------------------|------------------|-----|-------------------------|---------|
| | | | Pipe: 8 ft. | 3 in. coated leng | gth×0.310 in. I. | D. | | |
| 1 | 4450 | 31.5 | 0.00419 | 0.0321 | 13.06 | 320 | 586 | 0.00782 |
| 2 | 5240 | 31.5 | 0.00350 | 0.0321 | 10.90 | 320 | 586 | 0.00638 |

TABLE 4.—MASS TRANSFER OF SUCCINIC ACID FROM A WALL TO A FALLING WATER FILM

Tower: 34.5 in. coated length \times 0.807 in. I. D.

| Run | $4\Gamma/\mu$ | Avg. water temp. °C. | Normality exit water | Normality saturated solution | % Approach | Molecular diffusivity (lit. value) D_L , sq. ft./hr. | Apparent diffusivity (Nusselt Eq.), sq. ft./hr.×10 ⁵ | Wt. % wetting agent added to water |
|-----|---------------|----------------------------|----------------------------|------------------------------------|---------------|--|--|--|
| 1 | 1730 | 19.3 | 0.1114 | 1.12 | 9.94 | 3.1×10^{-5} | 25.7 | 0.05 |
| 2 | 1200 | 19.5 | 0.1264 | 1.12 | 11.3 | 3.1×10^{-5} | 19.2 | 0.05 |
| ્ર | 848 | 19.5 | 0.1826 | 1.12 | 16.3 | 3.1×10^{-5} | 20.9 | 0.05 |
| 4 | 481 | 19.5 | 0.2406 | 1.12 | 21.45 | 3.1×10^{-5} | 14.7 | 0.05 |

TABLE 5.—MASS TRANSFER OF ADIPIC ACID FROM A WALL TO A FALLING WATER FILM

Tower: 38.5 in. coated length \times 1.13 in. I. D.

| Run | $4\Gamma/\mu$ | Avg. water temp., °C. | Normality exit water | Normality saturated solution | % Approach | Molecular diffusivity*, D_L -sq. ft./hr. | Apparent diffusivity (Nusselt Eq.) sq. ft./hr.×10 ⁵ | Wt. % wetting agent added to water |
|-----|---------------|-----------------------------|----------------------------|------------------------------------|---------------|--|---|---|
| 1 | 1250 | 24.8 | 0.0343 | 0.2603 | 13.2 | 2.95×10^{-5} | 19.5 | 0.05 |
| 2 | 1373 | 24.8 | 0.0353 | 0.2603 | 13.6 | 2.59×10^{-5} | 22.6 | 0.05 |
| 3 | 1423 | 24.6 | 0.0363 | 0.2603 | 14.0 | 2.59×10^{-5} | 24.2 | 0.05 |
| 4 | 1530 | 24.5 | 0.0340 | 0.2603 | 13.1 | 2.59×10^{-5} | 24.3 | 0.05 |
| 5 | 1680 | 24.5 | 0.0329 | 0.2603 | 12.65 | 2.59×10^{-5} | 26.3 | 0.05 |
| 6 | 1703 | 24.3 | 0.0332 | 0.2603 | 12.78 | 2.59×10^{-5} | 27.6 | 0.05 |
| | | | •. | | | | | |

^{*}Estimated by method of Wilke (27).

Tower: 30% in. coated length $\times 0.794$ in. I. D.

| Run | $4\Gamma/\mu$ | Average Liquid temp., °C. | Normality exit | Normality Saturated solution | % Approach | Molecular diffusivity \dagger , D_L , sq. ft./hr. | diffusivity (Nusselt Eq.), sq. ft./hr.×10 ⁵ |
|-------------|---------------|---------------------------------|-------------------|------------------------------------|---------------|---|--|
| 1 | 2700 | 24.8 | 0.00162 | 0.0458 | 3.54 | $2.48{	imes}10^{-5}$ | 26.4 |
| $\tilde{2}$ | 2340 | 24.7 | 0.00168 | 0.0458 | 3.68 | 2.48×10^{-5} | 22.7 |
| $\bar{3}$ | 1980 | 24.7 | 0.00191 | 0.0458 | 4.16 | 2.48×10^{-5} | 21.7 |
| $\bar{4}$ | 1675 | 24.7 | 0.00194 | 0.0458 | 4.24 | 2.48×10^{-5} | 18.3 |
| 5 | 1350 | 24.5 | 0.00234 | 0.0458 | 5.10 | 2.48×10^{-5} | 17.1 |
| 6 | 1070 | 24.7 | 0.00288 | 0.0458 | 6.28 | $2.48{	imes}10^{-5}$ | 16.8 |
| 7 | 868 | 24.7 | 0.00354 | 0.0458 | 7.72 | $2.48{	imes}10^{-5}$ | 17.0 |
| 8 | 665 | 24.7 | 0.00454 | 0.0458 | 9.91 | 2.48×10^{-5} | 17.0 |
| 9 | 478 | 24.7 | 0.00588 | 0.0458 | 12.8 | 2.48×10^{-5} | 16.2 |
| 10 | 308 | 24.7 | 0.00837 | 0.0458 | 18.3 | 2.48×10^{-5} | 13.2 |

^{*} Ethanol used was 2B alcohol.

TABLE 7.-MASS TRANSFER OF PARAFFIN* WAX FROM A WALL TO A FALLING n-BUTANOL FILM

Tower: 35 in. coated length \times 0.772 in. I. D.

| Run | $4\Gamma/\mu$ | Liquid temp. °C. | Wt. % paraffin in exit liquid | Wt. % paraffin in saturated solution | % Approach | $\begin{array}{c} \text{Molecular} \\ \text{diffusivity} \dagger \\ D_L\text{-sq. ft./hr.} \end{array}$ | Apparent diffusivity (Nusselt Eq.), sq. ft./hr.×10 ⁵ |
|-----|---------------|---------------------|--|---|---------------|--|---|
| 1 | 376 | 38.3 | 0.124 | 1.50 | 8.27 | $0.503\times10^{-5}\ 0.503\times10^{-5}\ 0.503\times10^{-5}\ 0.503\times10^{-5}\ 0.503\times10^{-5}\ 0.503\times10^{-5}$ | 10.7 |
| 2 | 792 | 38.1 | 0.0718 | 1.50 | 4.79 | | 12.3 |
| 3 | 903 | 38.0 | 0.0676 | 1.50 | 4.51 | | 13.2 |
| 4 | 955 | 38.2 | 0.0655 | 1.50 | 4.37 | | 13.4 |
| 5 | 1220 | 38.0 | 0.0669 | 1.50 | 4.45 | | 19.2 |

^{*} B. P. of paraffins correspond to C23. † Estimated acid to method of Wilke (27).

NOTATION

- a = interfacial area per unit liquid volume, sq.ft./cu.ft.
- B = total film thickness, ft.
- $C_p = ext{specific heat}$ $C = ext{concentration of component in}$ liquid
- $C_l = initial mixed concentration at$ time zero or at top of tower
- $C_i = \text{equilibrium concentration at}$ interface
- $C_2 =$ mixed concentration in liquid, distance L from top of tower
- $\Delta C_m = \text{mean concentration driving}$ force
- D =tube diameter, ft.
- $D_L = \text{molecular}$ liquid diffusivity, sq.ft./hr.
- $E_a = apparent$ liquid diffusivity, sq.ft./hr.
- $E_D = \text{eddy liquid diffusivity, sq.ft.}/$
- G = mass velocity, lb./(sq.ft.)(hr.)
- g = acceleration due to gravity, ft./sec.2
- $g_c = \text{conversion factor, lb. mass/}$ (ft.) (lb. force) (sec. 2)
- $H_L = \text{height of liquid film transfer}$ unit, ft.
- $j_D = \text{mass}$ transfer factor, dimensionless
- k =thermal conductivity, P.cu./ (hr.) (sq.ft.) (°C./ft.)
- L =tube length, ft.
- P = lb. force/sq.ft.
- s = distance from gas-liquid interface to point in liquid film, ft.
- $T_o = \text{temperature of wall, °C.}$
- $T_1 = initial$ temperature at time

- zero or at the top of the tower
- $T_2 =$ liquid temperature, distance Lfrom top of tower
- V = velocity, ft./hr.
- $V_m = \text{maximum velocity, ft./hr.}$
- y =distance from wall to point in liquid film
- $\mu = \text{viscosity, lb./(ft.)(hr.)}$
- $\rho = density, lb./cu.ft.$
- $\theta = \text{time, hr.}$
- $\Gamma = \text{liquid flow per unit periphery,}$ lb./(hr.)(ft.)

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[†] Diffusivity from I. C. T. (13) for stearic acid in absolute alcohol.