Gas Phase Controlled Mass Transfer in Two Phase Annular Horizontal Flow

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Concurrent flow of liquid and gas through pipe lines has received the attention of a number of investigators in recent years. The majority have measured pressure drop and liquid holdup in two-phase flow, and several correlations have been proposed to estimate these quantities (*4, 5, 7, 12, 13, 14*). Such information is needed to design piping systems through which two fluid phases flow simultaneously. However a much more detailed understanding of two-phase flow hydrodynamics is needed to predict mass transfer between the two fluid phases.

Several different flow patterns can be produced by pumping liquid and gas concurrently through a horizontal pipe (*1, 13, 17*). The particular pattern obtained depends upon fluid velocities, physical properties of the gas and liquid, and pipe diameter. In annular flow the gas flows through the central region of the pipe carrying some liquid with it as entrained droplets, while the remaining liquid flows as an annular film adjacent to the inside perimeter of the pipe.

The present work is a study of gas-phase controlled mass transfer between the two fluid phases in annular flow in horizontal pipes. The purpose of the work was threefold: to measure rates of gas phase controlled mass transfer, to determine the mechanisms of mass transfer, and to obtain information about the fluid mechanics of two-phase annular flow.

THEORY

Figure 1 shows a cross section of two-phase annular flow in a horizontal pipe. The asymmetry of the liquid film is typical in horizontal pipes, but the asymmetry decreases as the gas velocity increases. The average gas velocity is much higher than that of the liquid film, and to maintain annular flow the gas phase must be turbulent. The interface separating the gas phase from the annular film is not smooth, and the amplitude of interfacial waves has been measured (*15*). Entrained liquid droplets in annular flow probably result from the breakup of these waves with injection of the resulting droplets into the turbulent gas stream. Transport of liquid droplets into the gas stream and deposition of droplets carried by the gas stream onto the annular liquid film is defined as *interchange*. Except for interchange and the momentum transfer associated with interchange the gas-phase motion in annular flow is probably similar to flow through a pipe in which roughness of the pipe walls is analogous to the rough surface of the annular liquid film; that is flow of the gas phase near the annular film is similar to flow near the wall of a pipe through which a single fluid phase passes.

Transfer of solute between the two phases is intimately related to the hydrodynamics of two-phase annular flow. Near the interface separating the gas and annular liquid film radial transport of the solute in the gas phase occurs primarily by molecular diffusion. The rate of mass transfer resulting from diffusion through the gas phase and transfer across the interface can be estimated by extrapolating wetted wall column data to the higher gas rates obtained in two-phase annular flow. Mass transfer rates in wetted wall columns, expressed in terms of j_D factors, have been correlated with gas Reynolds number based upon superficial gas velocity (*16*), but no measurements of mass transfer rates in wetted wall columns have been made for the high gas velocities encountered in two-phase annular flow. A second mechanism of mass transfer is diffusion of the solute between the entrained liquid droplets and the gas stream which carries the droplets. Calculation of mass transfer rates between the droplets and the surrounding gas is very difficult. Not only the distribution of droplet sizes but also the contributions to mass transfer by the continual formation and destruction of gas phase boundary layers around the droplets as they are buffeted about in the turbulent gas stream must be known. In this work all entrained liquid droplets are assumed to reach equilibrium with the gas phase before being deposited on the annular liquid film. This assumption will be supported below. In addition to diffusional mechanisms the overall rate of mass transfer is greatly increased by interchange of liquid droplets between the annular liquid film and the gas phase. Interchange contributes to overall mass transfer when the solute concentration of droplets differs from that of the annular film.

In the following analysis the overall gas phase controlled mass transfer rate in two-phase annular flow is ex-

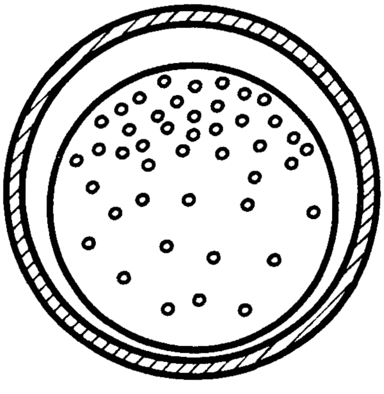

Fig. 1. Cross section of annular flow in a horizontal pipe.

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pressed quantitatively in terms of the mass transfer mechanisms described above. From measurements of solute concentration in the annular liquid film and in entrained liquid droplets the fraction of the total liquid flowing as entrained droplets can be calculated, and the rate of interchange of liquid droplets between the liquid annulus and the gas stream can be established.

Entrainment of Liquid Droplets

Entrainment is defined by the ratio L_e/L. This ratio can be calculated from solute concentrations in the annular liquid film and in the entrained liquid droplets. To demonstrate this a cross section of the pipe is considered; a mass balance of the total liquid flowing through the cross section is

$$L = L_a + L_e \tag{1}$$

The total molar flow rate of solute S through the cross section is

$$S = L_a x_a + L_e x_e + Vy \tag{2}$$

All molar flow rates defined above include solute. If the entrained liquid droplets are in equilibrium with a gas phase containing a low solute concentration, then

$$y = Hx_e/P \tag{3}$$

Combining Equations (1), (2), and (3) to eliminate L_a and y, and dividing through by L, one gets

$$\frac{L_e}{L} = \frac{S - Lx_a - VHx_e/P}{L(x_e - x_a)} \tag{4}$$

If the change in solute concentration in the gas and liquid streams is small, L and V at any cross section can be approximated by the gas and liquid flow rates at the inlet of the contactor. The total solute flow is usually known at the pipe inlet or can be calculated from an overall mass balance. Thus by measuring x_a, x_e, and P at one cross section sufficient information is available to calculate fraction entrainment from Equation (14).

Interchange of Liquid Droplets

Interchange rate, defined as the rate at which droplets are torn away from the annular liquid film and injected into the gas stream, has dimensions of mass/(time) (length of pipe). For fully developed steady state flow this rate must be equal to the rate at which droplets are deposited on the annular film. The following analysis shows how interchange rate I can be calculated from mass transfer measurements in two phase annular flow.

A solute balance written for a symmetrical annular liquid film in the differential length of pipe dl shown in Figure 2 is

$$L_a \frac{dx_a}{dl} = I\ (x_e - x_a) + K_G'P\ (y - y^*)\ 2\pi r \tag{5}$$

where

$$y^* = Hx_a/P \tag{6}$$

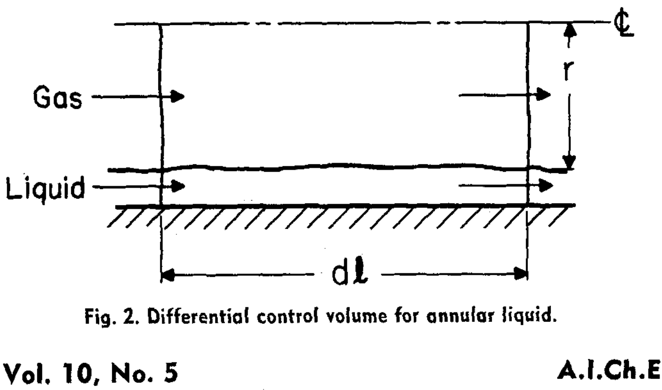

Fig. 2. Differential control volume for annular liquid.

The first term on the right of Equation (5) is the net rate of solute transfer to the liquid annulus by interchange. The second term is the net rate by diffusion across the gas-liquid interface of the liquid annulus. Equation (5) assumes that x_a is independent of radial and angular position in the annular film, and x_e is constant across the turbulent gas core. The assumption concerning x_a can be modified when variation of x_a with angular position has been determined. The distance r from the pipe center line to the annular film can be estimated from the Martinelli correlation (*14*) but is usually only slightly less than the pipe radius. Subtracting Equation (6) from Equation (3) one gets

$$P\ (y - y^*) = H\ (x_e - x_a) \tag{7}$$

Substituting Equation (7) into Equation (5) one obtains

$$\frac{dx_a}{dl} = \frac{I + 2\pi r K_G' H}{L_a}\ (x_e - x_a) \tag{8}$$

To solve this equation I is assumed independent of axial position. This is a valid assumption since as is shown later I is nearly independent of gas rate. Therefore the increased gas velocity associated with high pressure drop in two-phase flow does not have a large effect on interchange. After x_e is eliminated by use of Equation (4), integration of Equation (8) between axial positions l_1 and l_2, each located sufficiently far downstream that entrance effects are negligible, gives

$$I = \frac{(x_{a_2} - x_{e_1})}{(x_e - x_a)_{1m}}\ \frac{L_a}{l_2 - l_1} - 2\pi r H K_G' \tag{9}$$

A more convenient expression for the interchange rate can be obtained by relating K_G' to the Schmidt number and Reynolds number through the following equations:

$$j_D' = \frac{K_G' p_{BM}}{V}\ (N_{ScG})^{2/3}\ \pi \mathrm{r}^2 = \frac{f}{2} = 0.0395\ (N_{ReG})^{-1/4} \tag{10}$$

where

$$P_{BM} = \frac{(P_2 - Hx_{e_2}) - (P_1 - Hx_{e_1})}{\ln \dfrac{P_2 - Hx_{e_2}}{P_1 - Hx_{e_1}}}$$

The first equality in Equation (10) defines j_D' which correlates most wetted wall column data for gas phase controlled mass transfer. The second equality follows from the Reynolds analogy between mass and momentum transfer (*8, 9*), while the last equality is an empirical correlation of Fanning friction factor f with gas phase Reynolds number for single-phase flow through smooth pipes (*2*). This friction factor is useful in describing mass transfer between the gas and annular liquid film by diffusion only. The friction factor calculated from two-phase pressure drop is not suitable for use in Equation (10) because it includes momentum transfer due to interchange in addition to shearing stresses at the gas liquid interface. The last equality in Equation (10) which applies to flow through smooth pipes provides satisfactory correlation of gas phase controlled mass transfer in wetted wall columns (*16*), even though Emmert (*10*) has reported that waves are present on the gas-liquid interface of such columns. Substituting Equation (10) into Equation (9) one gets

$$I = \frac{(x_{a_2} - x_{a_1})}{(x_e - x_a)_{1m}}\ \frac{L_a}{l_2 - l_1} - (0.079)\ (N_{ReG})^{-1/4}\ (N_{ScG})^{-2/3}\ \frac{HV}{P_{BM} r} \tag{11}$$

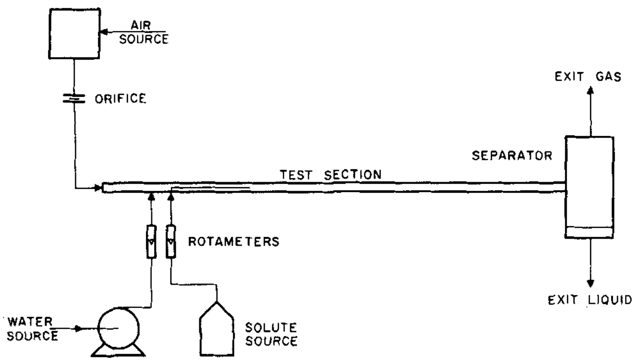

Fig. 3. Experimental equipment.

Overall Mass Transfer Calculations

The overall mass transfer coefficient K_G which includes mass transfer from both diffusion and interchange is defined by

$$K_G = \frac{(x_{a2} - x_{a1})\, L_a}{\Delta P_{1m} 2\pi r (l_2 - l_1)} \tag{12}$$

where

$$\Delta p_{1m} = H(x_e - x_a)_{1m} \tag{13}$$

The j factor, which includes the total mass transfer, is defined by

$$j_D = \frac{K_G p_{BM}}{V} (N_{ScG})^{2/3} \pi r^2 \tag{14}$$

Although mass transfer resulting from interchange is not dependent on N_{Sc}, j_D is used to correlate results because it takes pressure changes into account and simplifies the presentation.

EXPERIMENTAL MEASUREMENTS

For gas phase controlled mass transfer in two-phase annular flow Equations (4), (11), (12), and (14) are used to calculate entrainment, interchange, mass transfer coefficient, and j_D factor, provided that fluid flow rates are known and that the solute concentration in the annular liquid and entrained liquid is known at two points along the length of the contactor. The equipment and procedures used in this work are described in the following sections.

Experimental Equipment

Mass transfer rates in two phase annular flow were measured with the equipment shown in Figure 3. The test section was a horizontal tube 40 ft. long, constructed by joining 6-ft. sections of 1-in. I.D. extruded methacrylate tubing. Negligible flow disturbance resulted from the joints.

The entrance section, an extension of the test section, provided for introduction of liquid, gas, and solute into the test section. Water, the liquid phase, was pumped through a tee into the entrance section at right angles to the pipe axis. Air was saturated with water vapor and introduced at the inlet of the entrance section along the pipe axis. The solute, ammonia, passed through a ¼-in. O.D. steel tube mounted along the pipe's axis. The injection tube extended 31 in. downstream from the water inlet tee where the ammonia discharged into the test section and mixed with the air stream. Thus absorption of ammonia into the liquid phase did not begin until the annular flow pattern was established. The test section discharged into a 12-in. I.D. steel column which separated the gas and liquid phases without disturbing the upstream flow pattern.

Both annular and entrained liquid were sampled. Annular liquid was sampled through seven taps, each a 0.04-in. hole drilled through the top of the pipe. The first three taps were located at 3-ft. intervals beginning 3 ft. downstream from the ammonia discharge point. The remaining taps were 6 ft. intervals. The annular liquid sampling apparatus was a 30-cc. syringe connected by Tygon tubing to a 20-gauge hypodermic needle. To sample the annular liquid the needle was inserted through a tap sealing the sample tap into the annular liquid on the lower part of the pipe opposite the tap opening.

Entrained liquid droplets were sampled from two probes permanently installed in the test section. The first probe was 6 in. upstream from the first annular liquid sampling tap, and the other was 4 in. downstream from the last annular liquid tap. Each entrained liquid sampling probe was made of 0.07-in. O.D. steel tubing with one end protruding through the pipe wall and the other end facing upstream along the pipe's axis.

Seven pressure taps were spaced at 6-ft. intervals along the test section. The first tap was 11 in. downstream from the water inlet tee.

Experimental Procedure

Flow of water and air were adjusted to the desired rate. The flow pattern was observed through the transparent plastic test section, and pressure distribution along the contactor was measured. Ammonia was admitted to the contactor through the solute introduction pipe mounted along the axis of the entrance section. Ammonia rate was measured by rotameter and held constant during each run. Annular liquid samples were taken from the seven sample taps in the test section. Sample size varied from 5 to 15 ml. Approximately 10-ml. liquid samples were collected from the entrainment sampling probes at the same time annular liquid was withdrawn. The temperature of liquid leaving the separator was measured, and a sample of this liquid was obtained. Concentration of ammonium ion in each sample was determined by titrating with 0.1N hydrochloric acid with methyl red as an indicator.

RESULTS AND DISCUSSION

Entrainment

Measurements of solute concentration in the annular liquid and in entrained droplets at various axial positions in the pipe were used in Equation (4) to calculate the fraction of total liquid flowing as entrained droplets L_e/L. Equation (4) assumes that entrained droplets are in equilibrium with the surrounding gas phase. A comparison of results calculated from Equation (4) with direct measurements of entrainment tests this assumption. Entrainment in two-phase annular flow has been measured by Alves (*1*), Fritzlen (*11*), Budd (*3*), and Wicks and Dukler (*18*). They used probes to withdraw entrained liquid droplets from the pipe. Wicks and Dukler correlated entrainment data using a parameter $R = -(Q_L/Q_G)\,(N_{We_c})\,(E)/(dP/dl)_G$ and a parameter defined by Martinelli (*14*) as

$$X = [(dP/dl)_L/(dP/dl)_G]^{1/2}$$

Their correlation is shown in Figure 4. Wicks and Dukler report that the entrance section design has some influence

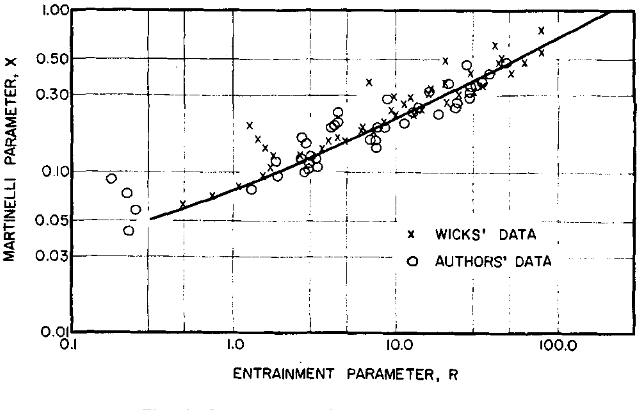

Fig. 4. Comparison of entrainment data.

on the amount of entrainment. Entrainment values calculated from Equation (4) are shown in Figure 4 with the data of Wicks and Dukler for the same type of entrainment section. The entrainment values calculated from mass transfer measurements agree with those data obtained by direct measurement. This supports the assumption made in deriving Equation (4), that entrained droplets are in equilibrium with the surrounding gas stream. Thus measurement of solute concentration in the annular liquid film and in entrained droplets for a gas phase controlled system is an alternative to the direct measurement of entrainment. Measuring liquid concentrations has the advantage of causing only very small disruption of the flow pattern as compared with direct measurement.

Mass Transfer

Although two-phase annular flow contactors are used for transferring solutes between gas and liquid phases, no mass transfer data for such contactors is available, therefore no rational basis for designing such equipment exists. For design purposes knowledge of the overall mass transfer coefficient K_G as a function of gas and liquid phase flow rates, fluid physical properties, and pipe diameter is needed. By measuring the composition of the annular liquid film and entrained liquid droplets at two different cross sections the overall mass transfer coefficient can be calculated from Equation (12). L_a in Equation (12) can be calculated from the fraction entrainment since $L_a = (1 - L_e/L)\ L$. The overall mass transfer coefficient is plotted in Figure 5 as a function of N_{ReG} with N_{ReL} as a parameter. Figure 5 indicates that the overall mass transfer coefficient increases if either liquid or gas flow rate is increased and is more strongly affected by liquid flow rate.

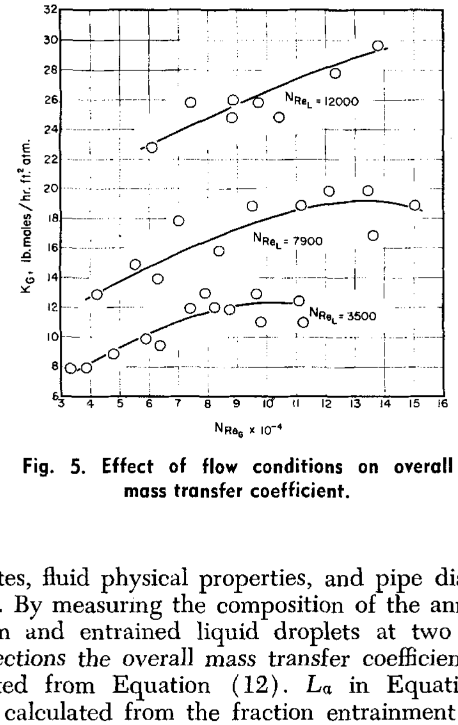

Fig. 5. Effect of flow conditions on overall mass transfer coefficient.

To separate the contributions to mass transfer by diffusion and interchange the mass transfer rates are expressed in terms of j_D factors. In this way the effect of pressure changes on mass transfer rates are taken into account. With Equation (14) used j_D factors were calculated from solute concentrations in the annular liquid film and in entrained droplets. The results are plotted in Figure 6 as a function of gas and liquid Reynolds number. Figure 6 is not intended to be a general correlation because effects of pipe size and fluid physical properties have not been investigated. Also shown in Figure 6 is an extrapolation of the $j_D = j_D' = f/2$ line which represents the value of the j_D factor expected if the only mechanism of mass transfer were gas phase controlled diffusion from the gas phase to the annular liquid film. The experimental data expressed as j_D factors lie above this line but approximately parallel to it for constant liquid flow rates. The difference between values of j_D and j_D' at any set of flow conditions is proportional to the quantity of solute transferred to the liquid annulus by diffusion between the gas and entrained liquid droplets with subsequent transport of entrained droplets into the annular liquid. Figure 6 shows that the difference between j_D and j_D' and thus the amount of solute transferred by interchange is a much stronger function of liquid flow rate than of gas flow rate. This can be explained by considering the interchange process in more detail.

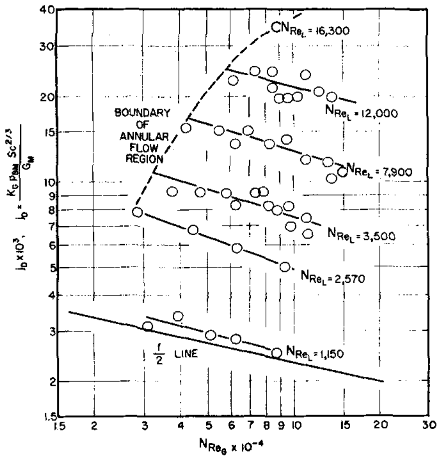

Fig. 6. Effect of flow conditions on j_D factor.

Interchange

The rate of interchange can be calculated from mass transfer measurements by Equation (9) or Equation (11). These results are shown in Figure 7. It is convenient to divide the rate of interchange I by the total liquid flow rate L and express the result as the fraction interchange or per cent interchange per foot of contactor length. Per cent interchange is shown as a function of gas and liquid Reynolds numbers in Figure 8. Interchange can be measured more directly by injecting liquid droplets containing a tracer directly into the gas stream at the pipe center line and measuring the tracer concentration in the annular film downstream from the point of injection. Experiments of this type were performed with a sodium chloride solution as the tracer. It is assumed that the motion of injected droplets is similar to that of drop-

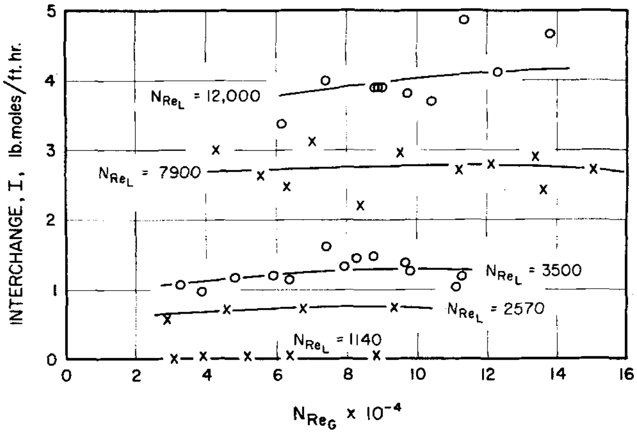

Fig. 7. Effect of flow conditions on interchange.

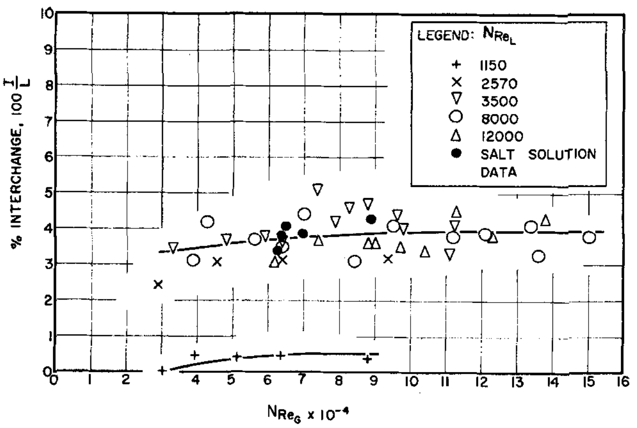

Fig. 8. Interchange as percent of liquid flow rate.

lets already present in the gas stream. Salt concentration of annular liquid and entrained droplets was determined by measuring the electrical conductivity of liquid samples removed through the various sample taps. Interchange was calculated from these data by using Equation (9). The last term in Equation (9) was omitted because diffusional transport between gas and liquid phases was absent. Results are shown in Figure 8. The agreement of interchange rate calculated from the two different types of experiments supports the hypothesis that the increase in mass transfer rates over those predicted by the $j_D = f/2$ line results from interchange.

Figure 8 shows that the per cent interchange per foot of contactor remains between 3 and 5% for N_{ReG} between 30,000 and 150,000 and for N_{ReL} between 2,750 and 12,000. However between N_{ReL} of 1,150 and 2,570 interchange drops to less than 0.5%. This can be explained in terms of the process which produces entrainment.

Interchange occurs when liquid is torn away from the annular film, transported by the gas stream, and eventually deposited on the liquid annular film. Interchange begins when waves on the surface of the annular film reach a sufficient amplitude to penetrate into the highly turbulent region of the gas phase. In this region the high shearing stress associated with high gas velocity breaks up the waves to form entrained liquid droplets. Determination of wave amplitudes in horizontal annular flow is complicated by asymmetry of the liquid film as shown in Figure 1. McManus (*15*) has shown that wave amplitudes are greatest in the region of maximum film thickness. McManus obtained data for horizontal annular flow in a 2-in. I.D. pipe and showed that wave amplitudes in the bottom of the pipe become equal to the thickness of the gas phase buffer layer at N_{ReL} 2,300. This result may differ for flow in a 1-in. I.D. pipe. However the fact that a sudden change in interchange occurs at $N_{ReL} = 2,400$, as shown in Figure 8, supports the argument that interchange begins when waves are first able to penetrate into the highly turbulent region of the gas stream. In vertical annular flow, as the liquid flow rate is increased, a point is reached at which there is a sudden increase in the quantity of entrainment, and at these flow conditions the wave amplitude on the gas-liquid interface of the annular film is approximately equal to the thickness of the gas-phase buffer layer (*6*).

If the hypothesis concerning onset of interchange is correct, then the rate of interchange will depend on the degree of penetration of waves into the highly turbulent part of the gas stream. This may explain the lack of dependence of per cent interchange on gas flow rate as shown in Figure 8. The hypothesis implies that per cent interchange is dependent on the ratio (wave amplitude)/(gas buffer layer thickness). Increasing gas flow rate decreases liquid film thickness, which in turn decreases wave amplitude. However the effect of smaller wave amplitude on interchange is compensated by the decrease in gas buffer-layer thickness with the result that changes in gas flow rate have little effect on rate of interchange.

SUMMARY

Rates of mass transfer in two-phase annular flow through a horizontal pipe have been measured. These data form a basis for the rational design of two-phase flow contactors when mass transfer rate is gas phase controlled.

Equations were derived for calculating entrainment from concentration measurements. Agreement of entrainment calculated in this manner with direct entrainment measurements is evidence of equilibrium between the gas phase and entrained liquid droplets. Calculation of entrainment from mass transfer measurements is an alternative to direct entrainment measurement.

Interchange of liquid between the annular film and entrained droplets was defined, and equations were derived to calculate this quantity from concentration measurements. Interchange was shown to have an important effect upon gas phase controlled mass transfer rates, and interchange rates were explained in terms of the fluid mechanics of two-phase annular flow.

NOTATION

D = pipe diameter, ft.
D_V = molecular diffusivity in gas, sq.ft./hr.
d = droplet diameter, ft.
$(dP/dL)_G$ = pressure gradient which would exist if gas flowed alone, lb. force/cu.ft.
$(dP/dL)_L$ = pressure gradient which would exist if liquid flowed alone, lb. force/cu.ft.
E = mass flow rate of entrained liquid, lb. mass/hr.
f = Fanning friction factor, dimensionless
H = Henry's law constant, atm.
I = interchange rate, lb. moles/(ft.)(hr.)
j_D = defined by Equation (14), dimensionless
j_D' = defined by Equation (10), dimensionless
K_G = overall mass transfer coefficient defined by Equation (12), lb. moles/(hr.)(sq.ft.)(atm.)
K_G' = mass transfer coefficient for transfer by diffusion only, lb. moles/(hr.)(sq.ft.)(atm.)
L = liquid molar flow rate, lb. moles/hr.
length coordinate of contactor, ft.
N_{ReG} = Reynolds number for gas phase = $(\rho_L U_L D/\mu)$, dimensionless
N_{ReL} = Reynolds number for liquid phase = $(\rho_L U_L D/\mu)$, dimensionless
N_{ScG} = Schmidt number for gas phase = $(\mu/\rho_G D_v)$, dimensionless
N_{Wec} = critical Weber number = 13, dimensionless
P = total pressure, atm.
P_{BM} = logarithmic mean of nondiffusing gas at phase boundary and in bulk of gas, atm.
Δp_{lm} = defined by Equation (13), atm.
Q = volumetric flow rate, cu.ft./hr.
r = radial distance from pipe center line to annular liquid film, ft.
R = entrainment correlation parameter = $-(Q_L/Q_G)(N_{Wec})(E)/(dP/dl)_G$, (lb. mass) (sq. ft.)/(lb. force)(hr.)
S = solute molar flow rate, moles/hr.
U = superficial fluid velocity, ft./hr.
V = gas molar flow rate, moles/hr.

X = Martinelli momentum transfer parameter $[(dP/dl)_L/(dP/dl)_G]^{1/2}$, dimensionless
x = mole fraction solute in liquid, dimensionless
y = mole fraction solute in gas, dimensionless
y^* = composition of vapor in equilibrium with annular liquid; defined by Equation (6), dimensionless
μ = fluid viscosity, lb. mass/(ft.)(hr.)
ρ = fluid density, lb. mass/cu.ft.

Subscripts

a = annular liquid film
e = entrained liquid
G = gas phase
L = liquid phase

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Longitudinal Dispersion of Thermal Energy Through Porous Media with a Flowing Fluid

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The rate of heat transfer in porous media with a flowing fluid present is determined by a combination of mechanisms. Broadly these may be grouped as bulk movement of the fluid, conduction in the solid and fluid phases, convective transfer of heat between the phases, convective eddy mixing or dispersion of the fluid phase in the porous-media interstices, and radiation.

A well-known example of the effect of these mechanisms in porous-media heat transfer is illustrated in Figure 1. A fluid at temperature T_o is flowing in one-dimensional, steady, piston flow through a homogeneous, stationary porous prism, also at temperature T_o. At the point $x = 0$ the temperature of the input fluid is suddenly changed to a new value T_i and held constant. A thermocouple, placed at $x = L$, will yield an S shaped response curve as the heat front arrives, not a step function as was introduced into the prism. Thermal energy will have dispersed in the direction of fluid flow and away from the step-function position, the amount of dispersion being determined by the heat transfer mechanisms which are important at the particular conditions of the experiment.

In the late 1920's Anzelius (*2*), Schumann (*27*), and others (*16, 23*) presented papers dealing with the mathematical description of heat flow by bulk fluid movement with convective transfer between solid and fluid phases. These works showed that as a consequence of the finite time required for heat transfer between the phases there was a dispersion of the energy giving rise to a characteristic S shaped temperature-response curve with a step-function input.

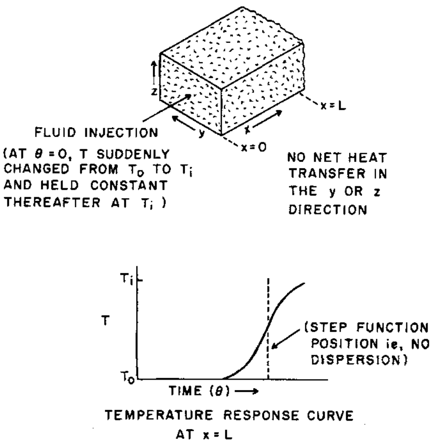

Fig. 1. Longitudinal heat transfer in porous media.

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