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MASS TRANSFER IN COCURRENT GAS-LIQUID FLOW

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Jon M. Heuss, C. Judson King, and Charles R. Wilke

August 7, 1964

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ABSTRACT

The absorption of ammonia and oxygen in horizontal cocurrent gas-liquid froth flow in a 1-in. I. D. pipe have been investigated. At superficial liquid rates between 2×10^6 and 3.2×10^6 lb/hr-ft² and superficial gas rates between 5×10^3 and 18×10^3 lb/hr-ft² the length of a transfer unit in both systems was between 0.5 and 4 feet. The effects of distance and temperature were also investigated. The absorption of ammonia was almost entirely gas phase controlled; the absorption of oxygen was almost entirely liquid phase controlled. The individual phase coefficients $k_L a$ and $k_G a$ are reported.

Using James and Silberman's data on bubble size and distribution in froth flow to estimate average bubble sizes, the ammonia absorption could be qualitatively and quantitatively explained by the model of unsteady state transfer from a stagnant sphere. Using an interfacial area estimate from the ammonia results, the oxygen absorption data were correlated by a Sherwood Number characterizing the transfer from the bubbles, a Reynolds Number characterizing the turbulence within the system, and the void fraction.

The void fraction was calculated from the Hughmark correlation. Pressure gradients were correlated by a modification of the Bankoff variable density homogeneous flow model.

I. INTRODUCTION

Gas absorption equipment, generally in the form of countercurrent packed towers or plate columns, is used extensively in the chemical industry. For the most part, investigations of mass transfer coefficients have been made in equipment utilizing countercurrent flow. However, cocurrent contacting of a gas and liquid seems entirely practical when a pure gas is used, when an irreversible chemical reaction is taking place in the liquid phase, and in general when equilibrium is not closely approached between exit streams. The phenomenon of flooding, which limits throughput in countercurrent operation, is not found in cocurrent operation. Thus there are a number of potential applications for contacting a gas and liquid cocurrently in a pipe.

The general problem of multiphase flow in a pipe has been extensively studied in recent years. Two phase flow is found in a growing number of chemical engineering processes, such as the production and transport of petroleum, heat removal from nuclear reactors, thermosiphon reboilers, and cryogenic processes.

Although the goal is a knowledge of the rates of momentum, heat and mass transfer in two phase flow systems, the vast majority of work thus far has been concerned with momentum and heat transfer, in particular with the macroscopic properties of pressure drop, holdup, and flow pattern. With the proper choice of transfer systems, individual phase coefficients may be inferred in a mass transfer study—something not possible in heat transfer.

The many variables involved complicate any approach to two phase flow problems. The recent reviews of Dukler and Wicks¹ and Scott² are quite complete and show the state of the art.

The complexity of two phase flow is dramatically brought out by the variety of possible flow patterns. The major patterns observed in horizontal two phase flow are shown in Fig. 1. They range from complete stratification of the gas and liquid, through mixtures of frothy slugs and plugs, to a froth of small bubbles dispersed in a continuous liquid phase. Although one cannot predict the flow pattern from basic

principles, empirical diagrams have been developed which allow such prediction. The most useful chart for predicting flow patterns in horizontal flow is that of Baker,³ shown in Fig. 2. . It is a plot of gas mass velocity G (in lb/hr-ft²) as a function of the ratio of liquid to gas mass velocities, L/G , with empirical correction factors Λ , for variations of density, and ψ , for variations of density, surface tension, and liquid viscosity, where

$$\Lambda = \left[\left(\frac{\rho_G}{0.075} \right) \left(\frac{62.3}{\rho_L} \right) \right]^{1/2} \quad (1)$$

and

$$\psi = \frac{73}{\sigma} \left[\mu_L \left(\frac{62.3}{\rho_L} \right)^2 \right]^{1/3} \quad (2)$$

The lines dividing flow patterns are actually broad transitions.

It is intuitively expected that highest interfacial areas and highest transfer rates will occur in the froth flow regime which is characterized by high throughputs and, hopefully, by a froth amenable to discussion using a homogeneous model. This study concerns mass transfer in horizontal cocurrent gas-liquid froth flow in a pipe.

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Fig. 1. Flow patterns in two phase gas-liquid horizontal flow.

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Fig. 2. Baker correlation of flow patterns in gas-liquid horizontal flow.

II. LITERATURE SURVEY

A. Froth Flow

Because of a lack of measurement techniques, there have been few studies of the more basic characteristics of turbulence, velocity distribution, interfacial structure, etc., in two phase flow. However, James and Silberman⁴ have reported data on bubble size and distribution in froth flow. They also report velocity distributions obtained with a pitot tube technique. Such measurements cannot be interpreted without information on the distribution and velocities of the two phases, the quantities they were measuring. By assuming a homogeneous froth, i.e., no slip, their results agreed with the universal velocity profile of single phase flow. They also found that the transition from slug to froth flow occurs essentially as predicted by Fig. 2. These results lend some experimental justification to the assumption of a homogeneous model to describe froth flow.

1. Pressure Drop

Many empirical and semiempirical correlations of pressure drop in multiphase flow have been presented.^{5,6} None are reliable over all of two phase flow; most are accurate within specific regions only. Models assuming homogeneous flow have been presented. They assume that the two-phase frictional pressure drop can be calculated as if an equivalent single phase mixture were flowing. One would expect that either froth or dispersed flow would be amenable to such a treatment.

Bankoff⁷ has introduced a variable density homogeneous flow model which assumes radial gradients in the concentration of bubbles which give rise to an overall slip even though no slip occurs on the microscopic scale. This explains how discrete bubbles in a presumably homogeneous froth can have a greater mean velocity than the liquid. Martin⁸ has applied the Hughmark⁹ correlation for void fraction (a modification of Bankoff's original equation) to Bankoff's viscous dissipation prediction and has included an acceleration term to successfully predict pressure drops in two and three phase froth flow. Martin has shown that the data of James and Silberman,⁴ Chisholm and Laird,¹⁰ and Johnson and Abou-Sabe¹¹ in froth flow are also successfully predicted.

2. Holdup or Void Fraction

It is an experimental fact that the in situ void fraction α (average void fraction across any cross section), is less than the void fraction in the feed ϕ , where

$$\phi = \frac{w_G/\rho_G}{w_G/\rho_G + w_L/\rho_L}$$

Some slip does occur. In this work α was from 0.8 to 0.9 of ϕ .

Dukler, Wicks, and Cleveland⁶ have tested the void fraction equations of Lockhart-Martinelli, Hughmark, and Hoogendoorn with a large set of experimental data, concluding that the Hughmark correlation is the best of the three.

B. Interfacial Area

James and Silberman present data on bubble size and distribution in froth flow at low void fractions. Sleicher¹² has measured the maximum drop size in the turbulent flow of two liquids at low holdup. Calderbank,¹³ Vermeulen, Williams and Langlois,¹⁴ and Rodger, Trice and Rushton¹⁵ have measured interfacial areas in liquid-liquid and gas-liquid dispersions in agitated tanks by light scattering techniques. Hinze¹⁶ and Kolmogoroff^{17,18} have derived equations predicting bubble size in locally isotropic turbulence.

Kolmogoroff's theory of locally isotropic turbulence hypothesizes that at high Reynolds Number the probability distributions defining turbulence are uniquely determined by ϵ , the energy dissipation per unit mass per unit time, and ν , the kinematic viscosity. Most flows are non-isotropic, but Kolmogoroff treated such systems by assuming that local isotropy exists if the volume under consideration is small compared to the main flow and large compared with η , the local scale of turbulence, where

$$\eta = \left(\frac{\nu^3}{\epsilon}\right)^{1/4}. \quad (3)$$

In this case the resulting probability distributions defining the turbulence depend on ϵ only.

Local isotropy has been applied to agitated tanks with encouraging results.^{19,20} One would expect that the assumptions of local isotropy might be met in a homogeneous froth and that it would be a powerful tool in predicting bubble sizes and turbulence levels in froth flow.

C. Mass Transfer in Two Phase Flow

Anderson²¹ absorbed ammonia in water in horizontal annular flow. He showed that two mechanisms contributed to the transfer. Diffusion from the liquid film as normally found in wetted-wall columns was one. The other was a continual interchange or exchange between gas and liquid, the gas stripping off liquid droplets which came to equilibrium in the gas stream and were eventually redeposited on the liquid film. The percentage interchange of liquid was essentially constant for gas rates in the range of superficial Reynolds Number Re_{GS} 30,000 to 150,000 and liquid rates in the range of superficial Reynolds Number Re_{LS} 1,150 to 2,570. Collins²² absorbed CO_2 in water in vertical annular flow at liquid rates from Re_{LS} 240 to 1,700 with negligible entrainment. Varlamov, et al.²³ studied the absorption of nitrogen oxides in NaOH in gas lift pumps of varying designs.

Scott and Hyaduk²⁴ absorbed pure CO_2 in horizontal tubes in a variety of flow patterns. In slug and annular flow the length of a transfer unit ranged from five to eighty feet, was inversely proportional to G , and was only slightly dependent on L and the tube size.

D. Mass Transfer to Interfaces

Harriott²⁵ reviews theories of mass transfer to rigid and mobile interfaces. Although the behavior of dispersed bubbles may depend considerably on the fraction of the volume which they occupy, previous work has generally neglected interference effects and has been applied to the motion of and rates of transfer to individual particles.

Very small bubbles rising in a fluid can be treated as rigid spheres up to diameters of about 0.2 cm.^{20,21,27,28,29} Transfer from the surface of drops in a moving stream with practically no relative velocity between the drops and surrounding fluid has shown rates analogous to those of stationary drops suspended in a stagnant fluid.^{30,31} In conditions of appreciable relative velocity the equation of Ranz and Marshall³² has been found to apply.

$$Sh = 2.0 + 0.60(Re)^{0.5}(Sc)^{0.33} \quad (4)$$

where the Sherwood Number is based on the drop size and the Reynolds Number is based on the drop size and the relative velocity.

Kronig and Brink³³ made a theoretical study of mass transfer from inside a circulating drop to the outer surface, finding that circulation increased the transfer rate 2.5 times. Handloss and Baron³⁴ derived an equation assuming transfer within the drop was entirely by eddy diffusion.

III. EQUIPMENT

The equipment is shown schematically in Fig. 3. Figure 4 is a photograph of the equipment. Air and water were brought together in a mixing section to form a froth which passed through a 1-in. I. D. Lucite test section into an aluminum cyclone separator. The liquid fraction from the separator was returned to a liquid storage tank for recirculation while the gas fraction passed out into the atmosphere.

The liquid was pumped from a 55 gal. steel storage drum and metered into the mixing section. The air supply was a 100 psig dry air line. The air, reduced by a regulator system, was heated, filtered, metered and delivered to the mixing section. The mixing section, as shown in Fig. 5, was made from standard galvanized pipe. The air passed through a 1-in. pipe; the liquid passed through the annulus between the 1-in. pipe and a concentric 2-in. pipe. A cap on the end of the 1-in. pipe was turned down and drilled with 32 1/16-in. diam. holes. The bubbles were formed as the air passed through the cap and were mixed with the liquid in a 2-in. x 1-in. reducer. The 4 ft. long Lucite test section had pressure and sampling taps drilled and tapped (1/8-in. N. P. T.) at points 6-in., 12-in., 18-in., and 42-in. downstream so that measurements could be made over 6-in., 1 ft., and 2 ft. distances.

For oxygen absorption, liquid samples were drained from the test section and metered through polarographic oxygen cells. For ammonia absorption, liquid samples were drained from the test section and metered through electrical conductivity cells. A detailed description of the equipment, the sampling and measuring devices in particular, will be found in the Appendix.

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Fig. 3. Schematic drawing of gas-liquid contacting apparatus.
 (P) pressure gauge; (T) thermometer; (S) sampling tap and measuring cell; (↑) rotameter.

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Fig. 4. Photograph of contacting apparatus.

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Fig. 5. Mixing section. (a) cross section. (b) plan view.

IV. PROCEDURE

A. Oxygen Absorption

First, the water circulation system was turned on. With only water flowing in the test section, valves on the sampling rotameters were opened, allowing water to pass through the two polarographic cells at a constant rate (1 to 2 cc/min). As the water circulated, it became saturated with air at one atm., as confirmed by an analysis by the Winkler method. After the cell signals came to steady state, the signals were recorded as the oxygen concentration in water in equilibrium with air at one atmosphere and the temperature of the water.

The sampling valves were then closed to keep air from entering the sampling cells while the air was turned on. Next, the air was turned on and the air and water flow rates were adjusted for the run. The sampling valves were opened and the flow rate of water through the cells was adjusted to be equal to that previously used to measure the oxygen concentration in water at one atm. At this flow rate no entrained bubbles entered the sampling line. The separator was open to the atmosphere so that the liquid returning to the storage tank was in equilibrium with air at one atmosphere and the temperature of the run (again as confirmed by the Winkler method). Since the temperature remained constant during a run, the concentration of oxygen in the liquid entering the test section was constant.

The liquid was contacted with air at a higher pressure in the test section and absorbed air; the equilibrium concentration at any point being determined by the pressure. The cell readings were recorded after 10 to 15 minutes at steady state. The liquid and gas rates, the temperatures of gas and liquid, the pressure drop between sampling taps, and the pressure at one of the sampling taps were also recorded. If any air was seen in the transparent polarographic cell housing the readings were disregarded and the run terminated. After each run, the system was again run with only water flowing to check the reference concentration before proceeding to another run.

B. Ammonia Absorption

The gas and liquid systems were turned on and set to the desired flow rates. In the ammonia system, three sampling taps and three conductivity cells were used—one for the liquid entering the mixing section and two in the test section. Valves on the three sampling lines were opened and liquid samples were metered at 2 cc/min. through each line. With the froth flowing, the ammonia regulator was opened and anhydrous ammonia was metered into the gas stream to give about 2% ammonia entering the mixing section. The system absorbed for two minutes and then the Hoke valves above and below each conductivity cell were simultaneously shut off capturing three samples. The bulk concentration approximately doubled during the 120 seconds absorption was taking place. Since the samples were captured within a two or three second period, the bulk concentration changed at most 3 percent during the sampling period. After the ammonia was shut off, conductivities of the three samples were read and recorded along with the temperatures of gas and liquid; flow rates of gas, liquid and ammonia; pressure drop between taps, and the pressure at one of the taps.

V. EXPERIMENTAL PROGRAM

A series of preliminary experiments was carried out to insure that the flow within the test section was fully developed. These included visual observations, high speed photographs, and pressure drop measurements. A number of simple mixing sections were tried. The final configuration (shown in Fig. 5) provided for the formation of bubbles and their mixing with the liquid stream. It also provided the most stable operation (least slugging) and developed the energy dissipation rates within a short distance.

A series of experiments was then carried out to investigate the effects of liquid rate, gas rate, temperature, and distance on the absorption of ammonia in water. The runs were all made near room temperature with distilled water and approximately 2 mole percent NH_3 in the entering air stream. The same variables were investigated in a series of experiments on the absorption of air (the rate of oxygen absorption being measured and reported herein).

VI. TREATMENT OF DATA

Overall mass transfer coefficients K_G and K_L are defined by the equations

$$N_A = K_G (p - p_e) \quad (5)$$

$$N_A = K_L (c_e - c) \quad (6)$$

where N_A is the flux in lb moles absorbed per unit time per unit area, p_e is the partial pressure of solute over a solution having the composition c , and c_e is the concentration of a solution in equilibrium with the solute partial pressure p . Since the interfacial area per unit volume, a , is not known, one can write

$$N_A a \, dl = K_G a (p - p_e) \, dl = K_L a (c_e - c) \, dl \quad (7)$$

where l is the length of test section.

For the measurement of the absorption of oxygen and ammonia, the flow of the bulk gas and liquid phases was essentially constant, so that the material balance can be approximated by

$$L_M dx = -G_M dy = N_A a \, dl. \quad (8)$$

Substituting (8) in (7), rearranging, and integrating with K_G and K_L assumed constant yields

$$l = - \frac{G_M}{K_G a P} \int_{p_1}^{p_2} \frac{dp}{p - p_e} \quad (9)$$

and

$$l = \frac{L_M}{K_L a P_M} \int_{c_1}^{c_2} \frac{dc}{c_e - c} \quad (10)$$

Following Chilton and Colburn³⁵ the integrals (9) and (10) are defined as "number of transfer units"

$$(NTU)_{OG} = \int_{p_2}^{p_1} \frac{dp}{p-p_e} \quad (11)$$

$$(NTU)_{OL} = \int_{c_1}^{c_2} \frac{dc}{c_e - c} \quad (12)$$

The "length of a transfer unit" is

$$(LTU)_{OG} = \frac{G_M}{K_G a P} \quad (13)$$

$$(LTU)_{OL} = \frac{L_M}{\rho_M K_L a} \quad (14)$$

Following Sherwood and Pigford³⁶,

$$(LTU)_{OG} = (LTU)_G + \left[\frac{mG_M}{L_M} \right] (LTU)_L \quad (15)$$

and

$$(LTU)_{OL} = (LTU)_L + \left[\frac{L_M}{mG_M} \right] (LTU)_G \quad (16)$$

where $(LTU)_G$ and $(LTU)_L$ are defined as

$$(LTU)_G = \frac{G_M}{k_G a P} \quad (17)$$

$$(LTU)_L = \frac{L_M}{\rho_M k_L a} \quad (18)$$

where $k_L a$ and $k_G a$ are the coefficients defined on the basis of interfacial concentrations.

At the liquid and gas rates used, mG_M/L_M for ammonia was always less than 1/200 so the $(LTU)_{OG} = (LTU)_G$ and the transfer was almost completely gas phase controlled. Similarly L_M/mG_M for oxygen absorption

was always less than 1/50 so that $(LTU)_{OL} = (LTU)_L$ and this case was almost completely liquid phase controlled.

In reality Eqs. (15) and (16) are based upon the classical additivity of resistances concept which may not hold true because of the relatively wide range of bubble sizes to be expected, as shown in Fig. 21. However, the conclusions regarding gas and liquid phase control for the oxygen and ammonia systems should still be valid.³⁷

The integral (12) may be evaluated analytically if the driving force $c_e - c$ at any point is a linear function of the solute concentration at that point. If this is true (12) becomes

$$(NTU)_{OL} = \frac{c_2 - c_1}{(c_e - c)_1 - (c_e - c)_2} \ln \frac{(c_e - c)_1}{(c_e - c)_2} \quad (19)$$

However, if c_e is a constant, (12) integrates to

$$(NTU)_{OL} = \ln \frac{(c_e - c)_1}{(c_e - c)_2} \quad (20)$$

Similar conclusions may be made for Eq. (11).

A. Ammonia

The back pressure of ammonia³⁸ can be calculated from

$$\log_{10} p_e/s = 4.699 - 1922/T \quad (21)$$

where p_e is the equilibrium partial pressure of NH_3 , s is the molality of NH_3 g-moles/l, and T is the temperature in degrees Kelvin. The high ratio of liquid to gas rates used herein kept $s < 10^{-2}$. In this case p_e is always less than 10^{-4} atm. which was negligible compared to p . Therefore Eq. (11) integrates to.

$$(NTU)_{OG} = \ln [p_1/p_2] \quad (22)$$

This analysis assumes constant G_M and constant P . As ammonia is being absorbed water is being vaporized so the G_M is slightly changed. In the measurement section P changes about 5 percent while p changes by a factor of 2 or more. These effects were taken into consideration and small correction were applied. They changed $(NTU)_{OG}$ by about 5 percent which was approximately the precision of the conductivity measurements.

B. Oxygen

In the oxygen case, water saturated with air at one atm. and the temperature of the run was contacted with air at higher pressures with the temperature remaining constant. Since the Henry's Law constant of oxygen is independent of pressure up to about 10 atm., the equilibrium concentration of oxygen is given by

$$c_e = y_{O_2} (P - p_w) / H \quad (23)$$

where y_{O_2} is the mole fraction of oxygen in dry air, p_w is the vapor pressure of water, and H is the Henry's Law constant. In the present case, dry air was contacted with the water, so water was being vaporized while air was being absorbed by the water. The amount of water vaporized was enough to cool the liquid up to 6°F/hr. However, the water was heated by the pump approximately the same amount so that the temperature remained constant during a run. The Schmidt Numbers of H_2O and NH_3 in air are about equal. Therefore, the ammonia results were used to calculate p_w at any point.

The pressure was decreasing down the test section. Because of this the pressure drop was increasing slightly. However, over the one- and two-foot sections used, the assumption of P linear in distance was adequate. Within the measuring section, P changed at most 10 percent and dP/dl changed less than 5 percent, so that the assumption of linearity lead to an error of less than 1 percent, well within the accuracy of the experimental measurement of P .

The $(NTU)_{OL}$ integral may be discussed with reference to Figs. 6, 7, and 8. Figure 6 shows the oxygen concentration as a function of distance down the test section. The curve denoted by B is the equilibrium line: since the equilibrium concentration at any point is determined by the pressure. Points c_1 and c_2 represent the measured oxygen concentrations. Figure 7 depicts the driving force $c_e - c$ as a function of c . Figure 8 shows the curve $1/(c_e - c)$ as a function of c , the area under which would give the value of the $(NTU)_{OL}$ integral.

Because c_e is not constant and $c_e - c$ is not linear in c , Eq. (12) cannot be analytically integrated. However, we may look at two limiting cases.^{39,54} For case one we assume c_e to be a linear function of c rather than of distance. Now the curves denoted by A on Figs. 6, 7, and 8 apply and the (NTU) integral is given by Eq. (19). For the second case we assume c_e to be constant retaining the value c_{e1} throughout the test section. If we define the difference between c_{e2} and c_{e1} as δ , then c_{e2} (case 2) is equal to c_{e1} (case 1) + δ . Curves corresponding to this second case are labeled C on Figs. 6, 7, and 8. Once again $c_e - c$ is linear in c and the value of the (NTU) integral is given by Eq. (20) as

$$(NTU)_{OL} = \ln \frac{(c_{e1} - c_1)}{(c_{e2} + \delta - c_2)} = \ln \frac{(c_{e1} - c_1)}{(c_{e1} - c_2)} \quad (24)$$

where $c_{e2} + \delta - c_2$ is the fictitious driving force assumed to apply at the end of the test section.

On Fig. 6, the true equilibrium line B lies between the two limiting cases. It is closer to C at the beginning of the test section where c is changing rapidly with distance and approaches A near the end of the test section where c is changing less and less rapidly with distance. We want to know $c_e - c$ as a function of c when c_e is linear in distance. We can estimate this by the use of the following approximate equations derived from the two previous limiting cases.

The (NTU) integral is related to (LTU) by

$$(NTU) = \ell / (LTU) \quad (25)$$

MU-34450

Fig. 6. Concentration vs distance for oxygen absorption.

MU-34451

Fig. 7. Driving force vs concentration for oxygen absorption.

MU-34452

Fig. 8. $(NTU)_{OL}$ integral for oxygen absorption.

If we assume (LTU) to be constant, we can apply the criterion that $l/(NTU)$ is constant over any distance l^* within the measuring section so that

$$(LTU) = \frac{l_2 - l_1}{\int_1^2 \frac{dc}{c_e - c}} \quad (26)$$

and

$$(LTU) = \frac{l^* - l_1}{\int_1^{l^*} \frac{dc}{c_e - c}} \quad (27)$$

We have two equations which are limiting cases for the evaluation of the (NTU) integral. If we apply Eqs. (19) and (24) to evaluate the integrals of Eqs. (26) and (27) we arrive at:

$$\ln \frac{(c_e - c)_1}{(c_e - c)_{l^*}} = \frac{l^*}{l_2} \ln \frac{(c_e - c)_1}{(c_e - c)_2} \quad (28)$$

and

$$\frac{c_{l^*} - c_1}{(c_e - c)_1 - (c_e - c)_{l^*}} \ln \frac{(c_e - c)_1}{(c_e - c)_{l^*}} = \frac{l^*}{l_2 [(c_e - c)_1 - (c_e - c)_2]} \ln \frac{(c_e - c)_1}{(c_e - c)_2} \quad (29)$$

At any length l^* the value of c_e may be determined from the assumed linearity of c_e in length and a value of c may be determined by trial and error from either Eq. (28) or Eq. (29). Such calculations for a number of runs showed that both Eq. (28) and Eq. (29) give essentially the same curve on Fig. 6. This curve is marked B'. Curves derived from B and B' on Fig. 6 are denoted by B on Figs. 7 and 8.

The areas under the curves on Fig. 8 have been measured for a variety of runs including those with the largest and smallest corrections. The calculations showed that the value of (NTU) computed using Eq. (28)

is within 1 percent of that computed using Eq. (29). The calculations also showed that the area under B was closer to the area under A being less than the area under A by between 30 and 40 percent of the difference between the areas under A and C. The average empirical factor was 35 percent. Therefore the true $(NTU)_{OL}$ was taken as

$$(NTU)_{OL} = (NTU)_{OL}^A - 0.35 [(NTU)_{OL}^A - (NTU)_{OL}^C]. \quad (30)$$

The empirical correction $0.35 [(NTU)_{OL}^A - (NTU)_{OL}^C]$ averaged 0.08 transfer units, or about 10 percent of $(NTU)_{OL}^A$. The value of the (NTU) integral computed by Eq. (30) is within ± 0.01 transfer units of the value obtained by graphical integration. This precision is well within the reproducibility of the data.

VII. RESULTS AND DISCUSSION

The results of this investigation are presented in four parts. The first two parts are concerned with visual studies and pressure drop measurements made while the gas-liquid contacting apparatus was run over a wide range of flow rates. The third part is concerned with a study of the effects of liquid rate, gas rate, temperature, and distance on the absorption of ammonia in water. The last part is concerned with a study of the effects of the same variables on the absorption of oxygen.

A. Visual Studies

The mixing section always formed a froth. At flow rates below those of froth flow the froth broke up as it passed down the test section. It broke up into that flow pattern which was stable at the given conditions. As froth flow was approached from the slug flow region, fewer and fewer slugs were observable until the flow appeared to be homogeneous throughout the test section. The transition occurred as predicted by Baker—in agreement with the observations of Martin and James and Silberman.

High speed photographs were taken using a General Radio Strobotac. Slug flow appears to be a mass of small bubbles with many large slugs of gas as shown in Fig. 9. As shown in Fig. 10, froth or bubble flow is a mass of small bubbles. It is somewhat arbitrary where one defines the transition between these two types of flow since there are always a few slugs present in froth flow. Nevertheless, the area Baker calls froth or bubble flow satisfies a visual definition.

B. Pressure Drop

The hydrodynamic measurements of James and Silberman and visual observations indicate that froth flow is a homogeneous mixture of small bubbles. The assumptions of Bankoff's variable density homogeneous flow model as used by Martin would seem to be met in the present system.

1. Variable Density Model

Bankoff⁷ assumes that the gas-liquid mixture is a suspension of bubbles, the velocities of gas and liquid being equal at any point. However, there is a radial gradient in the concentration of bubbles, with the concentration of bubbles being a maximum at the center of the pipe and decreasing to zero at the wall. This gives rise to a radial distribution of void fraction (and therefore density) and an overall slip because the gas is concentrated in the high velocity region of the velocity profile. Bankoff assumes power law distributions for velocity and void fraction to derive an equation for the average void fraction α , which is given by

$$\alpha = K \phi \quad (31)$$

where

$$\phi = \frac{\frac{W_G}{\rho_G}}{W_G/\rho_G + W_L/\rho_L} \quad (32)$$

with the parameter K being only a function of the constants characterizing the distributions chosen. Martin⁸ shows that profiles modified to

ZN-4349

Fig. 9. Photograph of slug flow.

ZN-4350

Fig. 10. Photograph of froth flow.

include a central region of constant velocity and void fraction with power law distributions applying near the wall give the same equation, with K then being only a function of the constants characterizing the modified profiles.

From experimental void fraction data, Hughmark⁹ used Eq. (31) to correlate K as a function of

$$Z = \frac{(Re)^{1/6} (Fr)^{1/8}}{(1 - \phi)^{1/4}} \quad (33)$$

where

$$Re = \frac{2R_p \left(\frac{W_G + W_L}{A} \right)}{[\mu_L (1 - \alpha) + \mu_G \alpha]} \quad (34)$$

and

$$Fr = \frac{[U_{LS} + U_{GS}]^2}{2gR_p} \quad (35)$$

The correlation is shown in Fig. 11.

Bankoff and Martin derive the mean velocity and mean density averaged over the cross section as

$$\rho_m = \rho_L (1 - \alpha) + \rho_G \alpha \quad (36)$$

and

$$U_m = U_{LS} + U_{GS} \quad (37)$$

From a momentum balance

$$\left(\frac{dP}{d\ell} \right)_{\text{total}} = \left(\frac{dP}{d\ell} \right)_{\text{fric}} + \left(\frac{dP}{d\ell} \right)_{\text{accel}} \quad (38)$$

where $(dP/d\ell)_{\text{fric}}$ is the frictional pressure gradient and $(dP/d\ell)_{\text{accel}}$ is the pressure gradient due to acceleration. Martin derives $(dP/d\ell)_{\text{accel}}$ as

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Fig. 11. Hughmark correlation of void fraction.

$$\left(\frac{dP}{d\ell}\right)_{\text{accel}} = \frac{-\rho_m U_m}{g_c} \frac{dU_m}{d\ell} \quad (39)$$

where $dU_m/d\ell = -U_{GS}/P (dP/d\ell)_{\text{total}}$ for a perfect gas.

$$\text{Therefore} \quad \left(\frac{dP}{d\ell}\right)_{\text{total}} = \frac{\left(\frac{dP}{d\ell}\right)_{\text{fric}}}{1 - \frac{\rho_m U_{GS}}{g_c P} U_m} \quad (40)$$

The frictional pressure gradient is given by

$$\left(\frac{dP}{d\ell}\right)_{\text{fric}} = \frac{2f}{R_P g_c} \left(\frac{1}{2} \rho_m U_m^2\right) \quad (41)$$

where the friction factor f is of the form

$$f = \frac{B}{\text{Re}^n} \quad (42)$$

where B and n are assumed to be constants which depend only of the pipe, and the Reynolds Number for the mixture is given by:

$$\text{Re} = \frac{2R_P \rho_m U_m}{\mu_m} \quad (43)$$

Previous homogeneous models have introduced various definitions of mixture density and viscosity. Bankoff and Martin use

$$\rho_m = \rho_L(1 - \alpha) + \rho_G \alpha \quad (44)$$

and

$$\mu_m = \mu_L \quad (45)$$

This definition of mixture density agrees with that of Dukler, Wicks and Cleveland⁵ and Woods.⁴⁰ Many definitions of mixture viscosity have been proposed. Woods⁴⁰ and White⁴¹ have proposed

$$\frac{1}{\mu_m} = \frac{X_v}{\mu_G} + \frac{(1 - X_v)}{\mu_L} \quad (46)$$

and

$$\mu_m = X_v \mu_G + (1 - X_v) \mu_L \quad (47)$$

respectively where X_v is the weight fraction of vapor, which was quite low in the present case. Dukler, Wicks and Cleveland propose

$$\mu_m = \mu_L(1 - \alpha) + \mu_G \alpha \quad (48)$$

Bankoff suggests the use of Eq. (48) when the bubbles are large and easily deformed. For minute bubbles, which act hydrodynamically as solid particles, Bankoff suggests that the mixture viscosity be given by the Einstein expression for the apparent viscosity of a suspension of solid spheres in a liquid:⁴²

$$\mu_m = \mu_L(1 + 2.5\alpha) \quad (49)$$

Since the viscosity enters into the equations to a small power, usually 0.3 or less, and there is disagreement whether μ_m increases or decreases with increasing void fraction, a reasonable approximation appears to be to set μ_m equal to the liquid viscosity.

Whereas previous homogeneous models have assumed that the gas and liquid travel at the same velocity, the variable density model proposes that the average velocity of the gas phase is greater than that of the liquid phase because the gas is concentrated in the region of higher velocity. For the mixture velocity, Bankoff and Martin propose a mean velocity averaged over the cross section,

$$U_m = U_{TP} = U_{LS} + U_{GS} \quad (50)$$

where U_{LS} is L/ρ_L and U_{GS} is G/ρ_G . It appears that a homogeneous or average velocity based on the mixture throughput and mixture density,

$$U_m = U_{hom} = \frac{L + G}{\rho_m} \quad (51)$$

would be a better characteristic velocity since almost all the mass and hence almost all the momentum are in the liquid phase and U_{hom} is more closely related to U_{LS} when U_{LS} and U_{GS} differ. The mixture velocity of Eq. (51) satisfies the criterion that the mixture density multiplied by the mixture velocity gives the total throughput. It will be shown later that this definition better characterizes the energy dissipation and mass transfer results.

If the gas and liquid fractions are considered separately, the velocities are increased to $U_{LS}/(1 - \alpha)$ and U_{GS}/α because the mass is flowing through a lessened cross section. If the new velocities are weighted by the percent of cross section they occupy,

$$U_m = \frac{U_{LS}}{1 - \alpha} (1 - \alpha) + \frac{U_{GS}}{\alpha} (\alpha) \quad (52)$$

which reduces to Eq. (50). Since $\rho_G \alpha$ and G are very small compared to $\rho_L(1 - \alpha)$ and L respectively, Eq. (51) can be approximated by $L/\rho_L(1 - \alpha)$ which is a definition of average liquid velocity as indicated in Eq. (52).

The pressure gradient for the flow of a gas-liquid mixture may now be calculated if the mass flow rates, temperature, and pressure are known. The void fraction is determined from the Hughmark correlation—Fig. 11. The mixture density is calculated from Eq. (44), and the mixture viscosity is given by Eq. (45). The mixture velocity is calculated from Eq. (51). The Reynolds Number is calculated from Eq. (43). The friction factor is taken from an experimentally determined plot of f vs Re for single phase flow. The frictional pressure gradient is calculated from Eq. (41) and the total pressure gradient from Eq. (40). This prediction, hereafter referred to as "the model," depends on an

empirical void fraction correlation and an empirical single-phase friction factor curve; however, it does not depend on any empirical parameters determined from two-phase pressure drop measurements.

2. Results

A total of 180 measurements of pressure gradient in froth flow from five investigations were predicted by the model. Table I describes the experimental conditions for each set of data. Figure 12 shows the location of the data on a Baker diagram. Note that a line of constant liquid rate on Fig. 12 would have a slope of -1. A reference to higher or lower gas rates on this figure assumes that the increase or decrease in G occurs with L held constant.

Martin predicted pressure gradients for the 180 points with the model as defined above except that he used Eq. (50) for mixture velocity. He found that the model worked well for all the data enclosed within curve 2 on Fig. 12. Table II summarizes Martin's findings for the 112 points enclosed within curve 2. At gas rates above curve 2 the model predicted from 20 to 100 percent too large a pressure gradient. Within curve 2, the acceleration pressure gradients were up to 25 percent of the total pressure gradient. Martin was able to extend his correlation to include the region enclosed by curve 3 at high temperatures since at higher gas rates the model was able to predict his 190°F. air-water data better than it was able to predict the room temperature data.

The data of this investigation, enclosed within curve 1, cover a range of flow rates both inside and outside the range of Martin's correlation. As shown in Table II, it is substantiated that Martin's correlation breaks down at higher gas rates.

Pressure gradients have also been predicted for the 180 points using the model as originally defined (Eq. (51) for mixture velocity). Table III summarizes the results for 165 points enclosed by curve 4 on Fig. 12. Figure 13 compares the predicted and measured pressure gradients for the data of this investigation. Some of these measurements were taken over the one foot length marked A on Fig. 14, some over the three foot

Table I. Description of data in horizontal gas-liquid froth flow

Investigator	Conditions	No. of Points
Present study	air-water at room temperature. 1" I.D. Lucite tube	60
Martin ⁸	air-water at room temperature. 1" I.D. copper tube	24
Martin ⁸	air-water at 190°F. 1" I.D. copper tube	29
Martin ⁸	air-oil at 120°F. 1" I.D. copper tube	39
James and Silberman ⁴	air-water at room temperature. 2-1/2" I.D. galvanized pipe	7
Chisholm and Laird ¹⁰	air-water at room temperature. 1" I.D. smooth tube	10
Johnson and Abou-Sabe ¹¹	air-water at room temperature. 1" I.D. brass tube	11

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Fig. 12. Baker diagram of pressure gradient data. 1, data of present study; 2, , Martin correlation at room temperature; 3, , Martin data at 190°F; 4, , Correlation of present study.

Table II. Summary of error analysis using Martin's correlation.

Description of data and number of points used (N)	Average ^a Deviation (a.d.) (percent)	Standard ^b Deviation (s.d.) (percent)
Present study air-water at room temp. (45)	81.8	55.0
Present study-points within range of Martin's correlation (18)	17.2	22.0
Martin air-water at room temp. (17)	1.1	8.1
Martin air-water at 190°F (24)	-6.0	9.6
Martin air-oil at 120°F (30)	8.0	8.7
James and Silberman Chisholm and Laird Johnson and Abou-Sabe air-water at room temp	(22) -3.9	11.1
^a a.d. = $\frac{\sum \text{err}}{N}$ where $\text{err} = \frac{(\frac{dP}{dl})_{\text{calc}} - (\frac{dP}{dl})_{\text{exptl}}}{(\frac{dP}{dl})_{\text{exptl}}}$ (100)		
^b s.d. = $\left[\frac{\sum \text{err}^2 - (\sum \text{err})^2/N}{N-1} \right]^{1/2}$		

length F, and some over the last foot of the three foot length, indicating that the flow pattern and energy dissipation rates become fully developed over a short distance. The model predicts too large a pressure gradient at gas rates above curve 4 and seems to predict too low in the range of lesser G. For 15 points above curve 4 (listed in Table IX) the predicted acceleration gradients are greater than the predicted frictional pressure gradients. When acceleration effects become predominant one would expect slip to occur. If slip occurs, the average liquid velocity would be less than predicted by Eq. (51) and the model would be expected to break down, predicting too large a pressure gradient. However, in the region above curve 4 the liquid rate is still high enough so that a phase-inversion to dispersed flow or the formation of a central core of gas (annular flow) does not occur. The region appears to be a very broad transition between the defined flow patterns. The model has an average deviation of -19 percent and a standard deviation (defined as the standard deviation from the average deviation) of 17.4 percent for the 165 points enclosed within curve 4. As discussed later, the ammonia results indicate that the bubbles act largely as rigid spheres which, following Einstein, suggests that $\mu_m/\mu_L > 1$. An empirical expression for mixture viscosity taking this effect into account would increase the predicted pressure gradients moving them closer to the measured values. The model might also be improved by the determination of an empirical relation between the friction factor predicted by the model and the value of f which when substituted into Eq. (41) would give the experimental pressure gradient.

A still more important conclusion is that within each set of data, the standard deviation is small, indicating that U_{hom} is a good characteristic velocity. The energy dissipation per unit mass and time may be computed

$$\epsilon = \frac{g_c \left(\frac{dP}{dl} \right)_{exptl} U_m}{\rho_m} \quad (53)$$

Table III. Summary of error analysis using correlation of present study.

Description of data and number of points used. (N)	Average Deviation (a.d.) (percent)	Standard Deviation (s.d.) (percent)
Present study air-water at room temp. (45)	-2.2	15.5
Martin air-water at room temp. (24)	-35.2	11.3
Martin air-water at 190°F (29)	-39.1	3.2
Martin air-oil at 120°F (39)	-10.6	2.8
James and Silberman Chisholm and Laird Johnson and Abou-Sabé (28) air-water at room temp.	-23.0	7.2
All (165)	-19.0	17.4

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Fig. 13. Pressure gradient correlation for data of present study.

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Fig. 14. Measurement sections. A, 1-ft. test section; B, 2-ft. test section; C, 6-in. test section; D, 6-in. test section; E, 6-in. entrance section; F, 3-ft. test section.

for the present data over the one foot section. Figure 15 is ϵ calculated with $U_m = U_{TP}$ as a function of U_{TP} : Figure 16 is ϵ calculated with $U_m = U_{hom}$ as a function of U_{hom} . The average liquid velocity is perceptibly better at characterizing ϵ . One could just as easily plot $(dP/d\ell)_{exptl}$ as a function of U_m since the mixture density is the same in each plot and velocity appears in both abscissa and ordinate. Plots of this sort are shown in Figs. 17 through 20 for Johnson and Abou-Sabe's¹¹ data and for Martin's room temperature air-water data. Again U_{hom} is the better at characterizing ϵ .

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Fig. 15. Energy dissipation vs U_{TP} for data of present study.

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Fig. 16. Energy dissipation vs U_{hom} for data of present study.

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Fig. 17. $(dP/dl)_{\text{exptl}}$ vs U_{TP} : data of Johnson and Abou-Sabe.

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Fig. 18. $(dP/dl)_{\text{exptl}}$ vs U_{hom} : data of Johnson and Abou-Sabe.

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Fig. 19. $(dP/dL)_{\text{exptl}}$ vs U_{TP} : data of Martin.

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Fig. 20. $(dP/dl)_{\text{exptl}}$ vs U_{hom} : data of Martin.

C. Ammonia Absorption

1. Liquid and Gas Rates

The absorption of ammonia over the one foot section marked A on Fig. 14 was measured in a set of 12 runs at 4 gas rates (from 5,250 to 12,000 lb/hr-ft²) and 3 liquid rates (from 2,050,000 to 3,000,000 lb/hr-ft²). The results are listed in Table IV. The correction for liquid phase resistance is negligible at the flow rates investigated; therefore (LTU)_G is reported.

A least squares regression analysis by digital computer fit the data to the form

$$(LTU)_G = \frac{a}{L^b G^c} \quad (54)$$

with the result

$$(LTU)_G = \frac{1.04 \times 10^6}{L^{0.25} G^{1.1}} \quad (55)$$

and

$$(NTU)_G = 9.65 \times 10^{-7} L^{0.25} G^{1.1} \quad (56)$$

2. k_Ga

Values of the individual phase mass transfer coefficient k_Ga were calculated from

$$k_G a = \frac{G_M}{P_{ave} \ell} (NTU)_G \quad (57)$$

The volumetric coefficient k_Ga is the product of an individual phase coefficient and the interfacial area per unit volume. If one can estimate the interfacial area, one can separate a from the volumetric coefficient and gain greater insight into the individual phase coefficients k_G and k_L.

For a system of many spherical particles consisting of N_i particles of diameter d_i in a given size interval dispersed in a medium, the mean surface-volume diameter is defined as

$$d_{sv} = \frac{\sum N_i d_i^3}{\sum N_i d_i^2}, \quad (58)$$

where the summation is over all size intervals. For a volume fraction of dispersed phase α , the interfacial area per unit volume is given by

$$a = \frac{6\alpha}{d_{sv}}. \quad (59)$$

The simplest mass transfer model which can be assumed is that of unsteady state transfer from a stagnant sphere with negligible external resistance. This problem has been solved in a series solution:²⁹

$$Y = \frac{p_o - p_f}{p_o - p_e} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-\frac{n^2 4\pi^2 Dt}{d^2}} \quad (60)$$

where p_o is the partial pressure within the bubble at time $t = 0$, p_e is the partial pressure in the bubble in equilibrium with the main body of liquid, p_f is the average partial pressure after some time t , D is the diffusivity, and d is the diameter of the bubble. Vermeulen⁴³ has shown that the exact solution can be approximated by

$$Y = 1 - e^{-\frac{4\pi^2 Dt}{d^2}} \quad (61)$$

for $Y > 0.4$.

The differential transfer rate from a bubble is defined as

$$k_c a (p - p_e) = -\frac{dp}{dt}. \quad (62)$$

Rearranging and integrating from p_o at $t = 0$ to p_f at $t = t$,

Table IV. Ammonia absorption results.

$L \times 10^{-6}$ (lb/hr-ft ²)	$G \times 10^{-3}$ (lb/hr-ft ²)	(NTU) _G corrected to (25°C)	(LTU) _G (ft)	$\frac{k_G a}{\left(\frac{\text{lb moles}}{\text{hr ft}^3 \text{ atm}} \right)}$	U_{nom} (ft/sec)	α	$0.016 U_{\text{nom}}^{3.2} \alpha^2$
2.06	7.50	0.86	1.16	200.	24.6	0.622	176.
2.06	9.86	0.97	1.03	294.	28.1	0.668	307.
2.06	11.98	1.35	0.74	483.	30.2	0.692	415.
2.06	5.25	0.49	2.04	82.5	20.5	0.548	77.
2.61	5.22	0.56	1.79	87.	22.9	0.486	85.5
2.61	8.00	0.93	1.08	216.	27.8	0.576	223.
2.61	10.07	1.02	0.98	286.	30.4	0.611	335.
2.61	12.01	1.35	0.74	442.	32.7	0.640	457.
2.98	5.70	0.60	1.67	96.5	25.2	0.465	105.
2.98	7.82	0.96	1.04	205.	28.3	0.525	194.
2.98	10.50	1.14	0.88	310.	31.8	0.577	339.
2.98	11.82	1.54	0.65	465.	33.8	0.601	447.

$$p_o \int_{p_o}^{p_f} \frac{dp}{p-p_e} = \int_0^t -k_c a dt \quad (63)$$

for $p_e = 0$,

$$\ln \frac{p_f}{p_o} = -k_c a t. \quad (64)$$

If

$$\frac{p_o - p_f}{p_o - p_e} = Y \quad (65)$$

then

$$1 - Y = \frac{p_f}{p_o} \quad (66)$$

or

$$\ln (1-Y) = -k_c a t \quad (67)$$

or

$$Y = 1 - e^{-k_c a t} \quad (68)$$

Comparing Eq. (61) with Eq. (68) it is noted that

$$k_c a = \frac{4\pi^2 D}{d^2} \quad (69)$$

where $k_c a$ is in reciprocal seconds. Equation (69) is for an individual bubble. Introducing $k_c = k_G RT$, and changing from a single bubble to a system of volume fraction α consisting of many bubbles of uniform diameter d , Eq. (69) becomes

$$k_G a = \frac{\alpha 4\pi^2 D}{RT d^2} \quad (70)$$

However, there exists a distribution of bubble sizes. The percent equilibration is a volumetric average over all bubbles:

$$Y_{ave} = \frac{\sum Y_i V_i}{\sum V_i} = \frac{\sum Y_i N_i d_i^3}{\sum N_i d_i^3} ; \quad (71)$$

If the unsteady state diffusion model applies, the percent equilibration for an individual bubble is given by

$$Y_i = 1 - e^{-(NTU)_{Gi}} \quad (72)$$

where

$$(NTU)_{Gi} \sim \frac{1}{d_i^2} \quad (73)$$

If the overall rate of transfer can be expressed in terms of an average bubble, d_{ave} , the gas phase coefficient in lb moles/hr-ft³-atm is given by:

$$k_G^a = \frac{\alpha 4\pi^2 D}{RT d_{ave}^2} \quad (74)$$

A knowledge of bubble size and distribution is necessary to evaluate d_{ave} and to test the simple model which has been presented.

James and Silberman⁴ report data on bubble size and distribution in froth flow at low void fractions obtained photographically. They found that bubble sizes at different positions were reducible to each other using perfect gas considerations and their reported distributions were so reduced to atmospheric conditions. Smooth curves drawn through a typical set of their data are shown in Figs. 21 to 23. Figure 21 is the cumulative number of bubbles as a function of bubble diameter. Figure 22 is the cumulative area vs bubble diameter. Figure 23 is the cumulative volume versus bubble diameter. Note that the area is evenly divided among all the bubbles and that one-half the volume is enclosed within only five percent of the bubbles.

Values of d_{sv} have been calculated for several of the James and Silberman distributions using Eq. (58). When these mean surface volume diameters are compared with the distributions from which they were calculated, it appears that d_{sv} is equivalent to a diameter

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Fig. 21. Cumulative number of bubbles: James and Silberman data.

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Fig. 22. Cumulative area: James and Silberman data.

MU-34465

Fig. 23. Cumulative volume: James and Silberman data.

exceeded by only 5 to 10 percent of the bubbles which is defined as d_{90} to d_{95} where d_{95} is the diameter exceeded by five percent of the bubbles.

Since the interfacial area is dependent on d_{sv} , it would be expected that d_{ave} should be not much different from d_{sv} . However, the distribution of bubble sizes introduces two effects which must be considered. The first is that small bubbles equilibrate faster. The driving force for the smallest bubbles approaches zero and they contribute less and less to the transfer as time goes on. This acts to raise d_{ave} at longer times as Marshall³⁰ has found in a calculation for a similar distribution of evaporating drops. Secondly, an average or effective k_G is based on the area over which transfer is taking place if the driving force is uniform.

$$k_{G \text{ ave}} = \frac{\sum k_{Gi} A_i}{\sum A_i} \quad (75)$$

If the definition of a is substituted in Eq. (70), it can be seen that k_G is itself inversely proportional to d . Since k_G is inversely proportional to d ,

$$k_{G \text{ ave}} \sim \frac{\sum \frac{1}{d_i} N_i d_i^2}{\sum N_i d_i^2} = \frac{\sum N_i d_i}{\sum N_i d_i^2} \quad (76)$$

which introduces a mean "diameter-surface" diameter, d_{ds} ,

$$d_{ds} = \frac{\sum N_i d_i^2}{\sum N_i d_i} \quad (77)$$

Calculations similar to those which showed d_{sv} to be equal to d_{90} to d_{95} showed that d_{ds} is equivalent to d_{80} to d_{85} in the James and Silberman distributions. Inclusion of the product $d_{ds} d_{sv}$ rather than d_{sv}^2 in Eq. (74) would tend to decrease d_{ave} .

Equation (71) has been applied to several of the James and Silberman distributions in order to determine the effect of the distribution on d_{ave} . A Y_i was chosen for one bubble size. Then Y_i 's for

all other bubbles within the distribution were calculated by Eq. (72) and (73). Y_{ave} was calculated from Eq. (71) and related to the bubble size which would give the equivalent Y_1 at the same elapsed time. This bubble size was then compared to the distribution from which it was derived.

It was found that for very small times (no bubbles over 50 percent equilibration) d_{ave} was equivalent to d_{90} as would be expected if d_{ave} were the geometric mean of d_{ds} and d_{sv} . As transfer proceeds, d_{ave} increases so that at $0.4 < Y < 0.9$ (the range of Y measured in the present mass transfer runs) d_{ave} is equivalent to a diameter exceeded by 4 to 6 percent of the bubbles. Therefore d_{ave} is taken to be equal to d_{95} .

The data of James and Silberman were taken under conditions slightly different from the conditions for the mass transfer runs of the present work. An estimation of d_{95} for the conditions of the mass transfer runs will now be discussed.

Figure 24 is a plot of d_{50} and d_{95} from the James and Silberman data (at the experimental conditions—not adjusted to atmospheric pressure) as a function of U_{hom} . The two points with U_{hom} less than 15 ft/sec are in the transition region between slug and froth flows. These two distributions differ from those at higher U_{hom} by having considerably more large bubbles. If these two points are excluded it can be seen that d_{50} varies as $U_{hom}^{-1.5}$ and d_{95} varies as approximately U_{hom}^{-1} .

Measurements of high speed photographs showed that d_{50} near the wall in the present system was in the range of 0.01" to 0.03", in general agreement with James and Silberman at equivalent values of U_{hom} . However, there may be a significant wall effect. As Kolmogoroff states,¹⁸ the local scale of turbulence varies across the cross section so that drops which move away from the center of the pipe break up to smaller diameters than those which stay near the center. Indeed, Sleicher¹² found that drops in a liquid-liquid system at low void fraction break up sooner near the wall. James and Silberman do not mention a wall effect in their air-water system.

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Fig. 24. Bubble diameter vs U_{hom} : James and Silberman data;
○, 2-1/2 in. pipe; ●, 4 in. pipe.

James and Silberman correlated their bubble size data for 2-1/2" I. D. and 4" I. D. pipes in terms of a dimensionless bubble diameter d' given by

$$d' = U_{LS} d_{atm} \sqrt{\frac{\rho_L}{\sigma 2 R_P}} \quad (78)$$

where d_{atm} is the bubble diameter at atmospheric conditions (14.7 psia and 70°F). Their plot of d_{atm} vs $\sqrt{2R_P}/U_{LS}$ is shown in Fig. 25. Since the range of data is small, and Figs. 24 and 25 are about equally satisfactory representations of the data, it is inconclusive whether or not there is an effect of pipe size.

There may also be an effect of void fraction on bubble diameter. Calderbank¹³ measured interfacial areas in gas-liquid mixtures in agitated tanks in which gas was being sparged through the liquid in an open tank. He found d_{sv} proportional to $\alpha^{1/2}$ for a case of considerable relative velocity between phases and called it a coalescence effect. Vermeulen, Williams, and Langlois¹⁴ measured interfacial areas in gas-liquid mixtures in closed tanks and found d_{sv} proportional to about $\alpha^{1/3}$ in a case with smaller relative velocity between phases. In the case of negligible relative velocity one might still expect a variation of d_{sv} with α . Over the small range of void fraction investigated, this effect might well be masked by the scatter of data. In relating bubble sizes between void fractions of 0.05 and 0.50 one might expect an increase in bubble size of about a factor of 2. A small amount of undetectable slugging would probably change the bubble size distribution, increasing d_{95} slightly.

From the James and Silberman distributions at atmospheric pressure and low void fraction ($\alpha < 0.1$) d_{95} would be 0.05" to 0.10" at the velocities used in the present mass transfer runs. (The data in Fig. 24 are not corrected to atmospheric pressure; those in Fig. 25 are.) At the pressures within the present system, d_{95} would be decreased to 0.04" to 0.09". At the void fractions investigated (0.4 to 0.7), d_{95} would be increased by a factor of from 1.5 to 2.5.

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Fig. 25. Bubble diameter vs $\frac{\sqrt{2R_p}}{U_{LS}}$: James and Silberman data;

O, 2-1/2 in. pipe; Δ, 4 in. pipe.

Slugging and/or a wall effect might increase d_{95} by a factor of from 1.0 to 1.2. This leads to an estimated range of d_{95} between 0.06" and 0.27" for the ammonia runs. Any higher estimate of d_{95} would not be consistent with the photographic determination of d_{50} .

Since Y was always greater than 0.4 in the one foot measuring section, Eq. (74) can be applied to the results. Average bubble sizes are calculated by substituting $D_{NH_3} = 0.78 \text{ ft}^2/\text{hr}^{1/4}$ and values of $k_G a$ and α into Eq. (74). The calculation shows d_{95} to vary from 0.12 to 0.27 in., in agreement with our previous estimation of 0.06 in. to 0.27 in. based on the James and Silberman data. Any turbulence, circulation, breakup, oscillation, or distortion would increase the transfer. The conclusion therefore is that the bubbles are acting largely as rigid spheres, the high molecular diffusivity overriding any breakup or turbulence which might be occurring. This conclusion is consistent with the assumption of no slip used in predicting pressure gradients. Small bubbles flowing along with a turbulent liquid would be expected to act as rigid spheres if there is no relative velocity.

As shown in Fig. 26, a least squares regression analysis fit $k_G a$ as

$$k_G a = 0.016 \alpha^2 U_{hom}^{3.2} \quad (79)$$

If $k_G a$ varies inversely with the diameter squared and the bubble diameter is a function of U_{hom} , one can conclude that

$$d_{95} \sim (\text{const.}) U_{hom}^{-1.6} \quad (80)$$

where the constant is determined from values of d_{95} calculated from Eq. (74) and from the corresponding values of U_{hom} . This agrees with the dependence of James and Silberman's d_{95}

$$d_{95} \sim U_{hom}^{-1.5} \quad (81)$$

MU-34468

Fig. 26. Least squares regression fit of ammonia absorption data.

and Vermeulen's¹⁴ finding that d_{sv} in gas-liquid mixtures in agitated tanks varies as velocity to the -1.5 power. Local isotropy applied to liquid-liquid dispersions¹⁹ predicts

$$d \sim \epsilon^{-2/5} \quad (82)$$

Since ϵ is proportional to U_{hom}^3 , Eq. (82) would predict d_{sv} proportional to $U_{hom}^{-1.2}$ which has been found by Vermeulen to be the case in liquid-liquid dispersions.

Equation (74) predicts a dependence of $k_G a$ on α to the first power instead of the second power. The exponent on α in Eq. (79) can be varied between 1.8 and 2.3 and the fit will be approximately the same since there is scatter to the data and α was only varied over a narrow range. As stated earlier any dependence of d_{95} on α should be positive which would tend to decrease the dependence of $k_G a$ on α . The power on α may therefore be anomalous or it may be possible that $k_G a$ is a function of α due to breakup and deformation at the higher void fractions where something similar to close packing is approached. A larger percentage of bubbles breaking up and reforming would increase $k_G a$ because in the initial period of transfer (until $Y > 0.4$) $k_G a$ predicted by the unsteady state diffusion model is higher than $k_G a$ predicted by the approximation (Eq. (74)).

3. Distance

In the initial period ($Y < 0.4$), the unsteady state diffusion model predicts $k_G a$ higher than $k_G a$ predicted by the approximation. Indeed $k_G a$'s measured over the six inch entrance section were about twice as large as those measured over the next one foot where Y was always > 0.4 . This agrees qualitatively with the predictions of Eq. (60). Furthermore, the exact series solution predicts $k_G a$'s slightly higher than the approximation in the range of Y between 0.4 and 0.6. Since $k_G a$ is inversely proportional to $(LTU)_G$, $(LTU)_G$ should be lower in the six inch section C than in the one foot section A. Five runs

were made with the transfer being measured over the six inch section C. As shown in Table V, $(LTU)_G$ measured over the six inch section averages 20 percent lower than $(LTU)_G$ measured over the one foot section in agreement with the above prediction.

The length of a transfer unit increased down the test section, changing most rapidly at first and approaching a constant value. Equilibrium was approached too fast to measure the transfer over more than a one foot section.

4. Temperature

Since the gas phase Schmidt Number is independent of temperature, gas phase transfer coefficients are generally insensitive to temperature. However, Dwyer and Dodge⁴⁵ and Haslam, Hershey and Kean⁴⁶ and others have found that k_G decreased slightly with increasing temperature in ammonia absorption in packed beds. This may be primarily an effect of temperature on interfacial area. The results of Dwyer and Dodge over a small temperature range at fixed flow rates can be empirically fit by the equation

$$(LTU)_G = \text{constant } e^{0.013(T-25)} \quad (83)$$

where T is in degree centigrade. From Eq. (83) it can be seen that $(LTU)_G$ varies as

$$(LTU)_G \sim e^{0.013T} \quad (84)$$

The effect of temperature on $(LTU)_G$ was investigated in three runs (runs 17 to 19 in Table X) at fixed weight flow rates over a range from 17.7 to 28.4°C. The results are shown in Fig. 27 and are empirically fit by the equation

$$(LTU)_G \sim e^{(+0.010 \pm 0.002)T} \quad (85)$$

Table V. Transfer over six inch section C at 25°C.

$L \times 10^{-6}$	$G \times 10^{-3}$	$(NTU)_G$	$(LTU)_G$	$(LTU)_G^a$	deviation (percent)
		for 6" sect. C		for 1' sect.	
2.06	7.27	0.48	1.04	1.31	26.0
2.61	7.77	0.55	0.91	1.12	24.1
2.06	9.88	0.635	0.79	0.93	17.7
2.61	5.50	0.37	1.35	1.64	21.5
2.06	11.50	0.68	0.735	0.81	10.2

^a Predicted by Eq. (55) for one foot section A.

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Fig. 27. Effect of temperature on $(\text{LTU})_G$.

The flow conditions in a packed bed and in cocurrent gas-liquid froth flow would not be expected to be similar. There is therefore no significance to the similarity of Eqs. (84) and (85).

An attempt to check whether the temperature dependence of Eq. (74) is consistent with the measured temperature dependence of $(LTU)_G$ was inconclusive because the lack of knowledge concerning d_{95} and α precludes reliable estimation of their temperature dependence.

D. Oxygen Absorption

1. Liquid and Gas Rates

The absorption of oxygen was measured over the one foot section A on Fig. 14 in a set of 28 runs at gas rates from 5,150 to 17,600 lb/hr-ft² and liquid rates from 2×10^6 to 3.2×10^6 lb/hr-ft². The results, corrected for gas phase resistance using Eq. (16), are listed in Table VI. The correction was never more than 2 percent. Within the set, there were a number of duplicate runs to check reproducibility. One set of flow rates was used for five runs of the above set and four runs investigating the effect of temperature. When corrected to 25°C seven of these runs clustered within ±10 percent of the mean while two were over 30 percent away from the mean. In a number of cases one run or one run out of a set of similar runs differed markedly from the best line through the data when plotted as $(NTU)_L$ vs one flow rate, the other flow rate held constant. A total of six such runs were rejected.

A least squares regression analysis fit the remaining data as

$$(NTU)_L = 3.0 \times 10^{-16} (L)^{1.65} (G)^{1.23} \quad (86)$$

or

$$(LTU)_L = 3.3 \times 10^{15} (L)^{-1.65} (G)^{-1.23} \quad (87)$$

Table VI. Oxygen absorption results over 1' test section corrected to 25°C.

Run No.	$L \times 10^{-6}$	$G \times 10^{-3}$	$(NTU)_L$	$(LTU)_L$
1	2.68	6.80	0.96	1.04 ^a
2	2.79	5.75	0.45	2.22
3	3.20	5.90	0.62	1.62
4	2.68	7.80	0.55	1.83
5	2.05	13.60	0.80	1.26
6	2.20	17.65	1.58	0.64
7	2.02	12.55	0.86	1.17
8	2.13	7.78	0.44	2.29
9	2.08	7.40	0.39	2.59
10	2.57	7.45	0.68	1.47
11	2.86	5.50	1.10	0.91 ^a
12	2.61	7.73	0.67	1.50
13	2.00	11.10	0.61	1.65
14	2.61	5.20	0.41	2.46
15	2.61	7.42	0.90	1.11 ^a
16	2.61	10.25	0.92	1.09
17	2.98	5.15	0.51	1.97
18	2.61	12.10	1.00	1.00
19	2.98	7.60	1.27	0.79 ^a
20	2.98	10.30	1.45	0.69 ^a
21	2.24	10.00	1.27	0.79 ^a
22	2.24	12.20	1.05	0.95
23	2.79	12.10	1.10	0.91
24	2.24	5.82	0.32	3.17
25	2.05	5.82	0.28	3.58
26	2.05	10.40	0.55	1.83
27	2.24	10.20	0.66	1.52
28	2.24	12.00	0.81	1.23

^aRejected when compared to runs at similar flow rates.

2. $\frac{k_L a}{L}$

Values of $k_L a$ were calculated from

$$k_L a = \frac{L_M (NTU)_L}{\rho_M \ell} \quad (88)$$

where ρ_M is the molar liquid density. In reality $k_L a$ is an average over all bubbles. In the ammonia case, the small gas bubbles equilibrated appreciably increasing the average bubble size as transfer proceeded. For the oxygen runs, calculation showed that the smallest gas bubbles do not equilibrate appreciably, therefore the driving force is uniform and the average bubble size does not change as transfer proceeds.

Dimensional Analysis leads to

$$Sh = f(Re, Sc, \alpha) \quad (89)$$

A least squares regression analysis fit $k_L a$ as

$$k_L a = (\text{const.}) U_{\text{hom}}^{4.1} \alpha^{-2} \quad (90)$$

Using the estimation of d_{sv} and d_{ds} from the ammonia results, we express a in terms of d_{sv} and rearrange Eq. (89) into dimensionless form as

$$\frac{k_L d_{ds}}{D_L} = 0.0316 \left(\frac{2R_P U_{\text{hom}} \rho_L}{\mu_L} \right)^{0.9} \left(\frac{1}{\alpha} \right)^3 \quad (91)$$

where $k_L d_{ds} / D_L$ is a Sherwood Number and $2R_P U_{\text{hom}} \rho_L / \mu_L$ is a Reynolds Number. The correlation is shown in Fig. 28 and the results listed in Table VII. d_{ds} is taken as equal to $0.8 d_{sv}$.

The Reynolds Number is based on the pipe diameter while the Sherwood Number is based on the bubble diameter, indicating that k_L is itself inversely proportional to d_{ds} . Previous work involving mass transfer to and from spheres has generally been concerned with either conditions of significant relative velocity between the phases where

Table VII. Correlation of oxygen absorption results

Run	$k_L a \times 10^{-3}$ (hr ⁻¹)	U_{nom} (ft/sec)	α	Sh	$0.0316 Re^{0.9} \alpha^{-3}$
2	20.0	24.6	0.485	14,750.	15,700.
3	31.4	26.5	0.453	19,550.	20,500.
4	23.3	28.0	0.566	9,800.	11,000.
5	26.0	31.3	0.705	6,130.	6,350.
6	55.8	37.1	0.731	7,250.	6,350.
7	27.6	30.8	0.703	6,830	6,300.
8	14.7	24.9	0.613	8,270.	7,820.
9	12.7	24.3	0.613	7,660.	7,700.
10	27.7	26.4	0.559	13,950.	10,950.
12	27.6	27.0	0.563	12,750.	10,900.
13	19.3	29.2	0.649	5,710.	6,350.
14	16.6	23.5	0.496	13,600.	14,100.
16	38.1	30.6	0.614	11,000.	9,450.
17	24.0	24.5	0.448	19,400.	19,800.
18	41.5	33.0	0.642	9,050.	8,750.
22	37.3	31.8	0.681	8,700.	7,120.
23	48.9	35.0	0.638	8,730.	9,450.
24	11.1	22.7	0.554	9,250.	9,800.
25	9.0	22.0	0.579	7,900.	8,300.
26	17.8	28.5	0.674	5,920.	6,650.
27	23.4	29.4	0.655	7,280.	7,450.
28	29.0	32.0	0.682	6,640.	7,150.

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Fig. 28. Correlation of oxygen absorption data.

correlation was in terms of a Reynolds Number based on that relative velocity, or with conditions of practically no relative velocity which are analogous to those of a stationary particle suspended in an infinite stagnant fluid. In the latter case

$$k_L = \frac{2D}{d} + \sqrt{\frac{D}{\pi t}} \quad (92)$$

where t is the time from the introduction of the particle into the fluid. For a distribution of bubbles,

$$k_{L \text{ ave}} = \frac{\sum k_{Li} A_i}{A_i} = \frac{2D \sum N_i \frac{1}{d_i} d_i^2}{\sum N_i d_i^2} + \sqrt{\frac{D}{\pi t}} \quad (93)$$

or

$$k_{L \text{ ave}} = \frac{2D}{d_{ds}} + \sqrt{\frac{D}{\pi t}} \quad (94)$$

or

$$\frac{k_{L \text{ ave}} d_{ds}}{D} = 2 + \frac{d_{ds}}{\sqrt{\pi D t}} \quad (95)$$

The pressure drop and ammonia absorption results were amenable to treatment assuming no relative velocity on the microscopic scale. If it is assumed that there is no relative velocity, Eq. (94) would be expected to apply in general form to a turbulent fluid even though the surrounding medium was not infinite provided D was replaced by a transport coefficient which would properly describe the enhancement of the diffusion process provided by the turbulence. Although a properly chosen effective eddy diffusivity might do this, present knowledge of the turbulent characteristics of two phase flow is so limited as to make prediction of an effective eddy diffusivity extremely difficult.

The calculated Sherwood Numbers are very large (6,000 to 20,000); the rate of oxygen absorption was from 10 to 100 times larger than predicted by Eq. (95); the percent saturation accomplished was quite large.

Thus the high turbulence level causes steady state to be reached very fast. At steady state the first term on the right side of Eq. (95) predominates over the transient term and k_L is inversely proportional to d_{ds} . Thus the form of the Sherwood Number is established.

A Reynolds Number based on the pipe diameter would be expected to characterize the turbulence of the flowing continuous liquid phase. The 0.9 power on the Reynolds Number is similar to that found in heat and mass transfer in forced convection near fixed interfaces.

The void fraction appears in Eq. (84) to the minus third power. The power on α can be changed ± 0.3 and the standard deviation of the correlation is still comparable to the accuracy of the measurements since the range of void fraction was small and α was calculated from a correlation which must be considered accurate only within ± 10 percent.

A possible explanation of the negative power on α may be offered in terms of a damping effect on the turbulence. Hinze⁴⁸ points out that interactions between particles become important as the concentration of particles is increased in a suspension of solids. Indeed as the maximum packing of particles is approached a severe damping of the turbulence is expected. If the bubbles are assumed to be rigid spheres, close packing would be approached in the range of void fractions investigated. Bagnold,⁴⁹ working with solid particles, noted a freezing of the turbulence as close packing was approached. He defines a "linear concentration," λ , as

$$\lambda = \frac{d}{z} \quad (96)$$

where z is the mean radial separation and d is the particle diameter. This linear concentration is a unique function of the void fraction. It increases quickly with α as close packing is approached and a corresponding damping of the turbulence would be expected.

The Sherwood Number should also be a function of the Schmidt Number. Ranz and Marshall³² and Friedlander⁵⁰ found the Sherwood Number to be proportional to $Sc^{1/3}$ for single spheres in cases of relative

velocity between phases. Calderbank and Jones⁵¹ obtained the same result for dispersions of ion exchange particles in a mixing vessel; Calderbank and Moo-Young²⁰ found the $1/3$ power to apply for small bubbles in gas-liquid dispersions in aerated mixing vessels. The data of Dobbins⁵² and Kilmer⁵³ for gas-liquid mixtures in stirred vessels at high turbulence, when adjusted to be consistent with a consensus of diffusivities,⁵⁴ indicate that k_L varies with D_L to powers between 0.5 and 0.75. In light of the above results it is apparent that the Schmidt group should enter the correlation of Eq. (91) to some power, probably between 0 and 0.5. The power cannot be fixed on the basis of temperature effect data (Sec. 4, below) because of a lack of knowledge of the effects of temperature on d_{ds} and α in Eq. (91).

3. Distance

In treating the data it has been assumed that $k_L a$ is independent of distance in well developed froth flow.

$(LTU)_L$ in the six inch entrance section E was about 50 to 100 percent higher than in the one foot section A. As shown in Table VIII, a set of six runs with the six inch measuring section D have values of $(LTU)_L$ which average 17.5 percent lower than the corresponding $(LTU)_L$ measured over the one foot section A. Also in Table VIII, a set of five runs over the two foot section B are compared with the results over the one foot section A. Measurements over the two foot section had a large uncertainty because equilibrium was closely approached at the end of the section. With this in mind, one can conclude that the length of a transfer unit over the two foot section is approximately equal to that over the one foot section within the precision of the measurements.

Thus $(LTU)_L$ decreases rapidly with distance approaching a constant value after the six inch entrance section. $k_L a$ therefore increases to a constant value. This can be explained as a phase mixing effect using the following physical argument.

Table VIII. Effect of distance on oxygen absorption

$L \times 10^{-6}$	$G \times 10^{-3}$	$(NTU)_L$	$(LTU)_L$ for 6" sect.	$(LTU)_L^c$ for 1 ft. sect.
2.68	7.65	0.43 ^a	1.16	1.34
2.68	7.65	0.46 ^a	1.09	1.34
2.06	7.65	0.29 ^a	1.72	2.08
2.68	10.20	0.55 ^a	0.91	0.95
2.80	7.65	0.55 ^a	0.91	1.28
2.98	7.65	0.46 ^a	1.09	1.09
			for 2' sect.	
2.24	5.81	0.87 ^b	2.30	2.57
2.61	7.73	1.58 ^b	1.27	1.38
2.20	6.55	0.97 ^b	2.06	2.28
2.05	7.90	0.97 ^b	2.06	2.08
1.71	8.23	0.63 ^b	3.18	2.62

^aTransfer measured over 6" section marked D on Fig. 14.
^bTransfer measured over 2' section marked B on Fig. 14.
^cTransfer predicted over 1' section A on Fig. 14.

The gas forms small bubbles as it comes through the cap on the one inch pipe in the mixing section (see Fig. 5). The liquid, however, is being mixed in from the sides by the reducer. Initially the gas is all in the form of small bubbles concentrated toward the center of the pipe and most of the liquid is not in close contact with the gas. As the mixture flows down the test section, radial mixing occurs and the flow pattern is developed within a few diameters. The effect of the gas being initially more efficiently mixed into two phase flow than the liquid is shown in Fig. 29. For simplicity, we assume that one half the liquid forms a froth with the gas immediately and that the other half is mixed into the liquid after one half a transfer unit has occurred within both phases of the froth. The results are as follows. In the liquid controlled case, the liquid in the froth initially goes one half of a transfer unit toward saturation and when it is mixed with the rest of the liquid, the total liquid has undergone approximately one quarter of a transfer unit.

$$\begin{aligned} 0.5(\text{TU})_{\text{OL}}(0.5) + 0.0(\text{TU})_{\text{OL}}(0.5) &= (39.4\% \text{sat})(0.5) + (0.0\% \text{sat})(0.5) \\ &= 19.7\% \text{ sat.} \\ &= 0.22 (\text{TU})_{\text{OL}} \end{aligned} \quad (97)$$

In the gas phase controlled case, after the froth has undergone one half of a transfer unit, the gas which is all contained in the froth has undergone one half of a transfer unit.

$$0.5(\text{TU})_{\text{OG}}(1.0) = 0.5(\text{TU})_{\text{OG}} \quad (98)$$

Thus under the given conditions, a fewer number of liquid phase transfer units are developed per unit length in the entry region than in the pipe beyond the entry region. Gas phase mass transfer on the other hand is similar in both regions.

Liquid controlled

Gas controlled

MU-34471

Fig. 29. Mixing section effect.

4. Temperature

The liquid phase Schmidt Number is highly dependent on temperature. Thus liquid phase coefficients are generally quite sensitive to temperature. For air-water systems in packed beds, the temperature coefficients for the liquid controlled oxygen, hydrogen, carbon dioxide and chlorine systems can all be represented by⁵⁴

$$(LTU)_L \sim e^{-0.020T} \quad (99)$$

The effect of temperature was investigated in four runs at fixed flow rates over the range of 19 to 34°C. The results are shown in Fig. 30 and are fit by the equation

$$(LTU)_L \sim e^{(-0.042 \pm 0.008)T} \quad (100)$$

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Fig. 30. Effect of temperature on $(\text{LTU})_L$.

VIII. CONCLUSIONS

High speed photographs show that froth flow is a mixture of small bubbles. It occurs, as predicted by Baker, at superficial liquid mass velocities above 1.8×10^6 lb/hr-ft². As Martin has shown, Bankoff's variable density homogeneous flow model can be combined with the Hughmark correlation for void fraction to predict pressure gradients in froth flow, up to gas mass velocities of about 7,000 lb/hr-ft². The range of the correlation may be extended up to about 14,000 lb/hr-ft² by the use of a mixture velocity, U_{hom} , based on the mass flow and mixture density. However, this still does not encompass all of froth flow. The region outside the limits of the correlation, which appears to be a broad transition between-froth flow and annular and dispersed flows, involves such high gas rates that acceleration effects become predominant and slip occurs. It was found that the homogeneous mixture velocity mentioned above can be used as a parameter to characterize the energy dissipation per unit mass and time.

The gas phase controlled ammonia absorption can be qualitatively and quantitatively explained using the simple model of transfer from a stagnant sphere. Average bubble sizes calculated from the ammonia data using this model are in agreement with a prediction made from the data of James and Silberman—the only available on bubble size and distribution in froth flow. Any model which would include circulation, turbulence or other effects would predict more transfer than was observed. The bubbles act largely as rigid spheres with no relative velocity between phases with the possible exception of an effect of breakup at high void fractions.

By using an estimate of interfacial area deduced from the ammonia results to separate k_L from k_{La} , the liquid phase controlled oxygen absorption may be correlated by a Sherwood Number characterizing the transfer from the bubbles and a Reynolds Number characterizing the turbulence within the system. A negative dependence on void fraction is explained as an effect of damping of the turbulence.

The length of a transfer unit in both systems is between 0.5 and 4 ft. At the high velocities used, equilibrium is approached within 0.1 second. The combination of a high interfacial area and a high turbulence level is believed to explain the rapid transfer.

The transfer rate increases faster than the throughput so that the length of a transfer unit decreases as the residence time decreases. The rate coefficient k_L or k_G is dependent on the bubble size. As the turbulence level is increased, k_L is increased. The average bubble size is also decreased which then further increases k_L and at the same time increases both the interfacial area and k_G .

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APPENDICES

A. Details of Equipment

1. Liquid System

The water was stored in a 55 gallon steel drum equipped with a sight glass and thermometer. It was pumped by a water-cooled Peerless Type PR centrifugal pump (rated at 60 G.P.M. at 100 ft. of head). The water flow rate was controlled by a 2" Globe valve in the bypass line. The output of the pump passed through a 1" check valve and a set of two rotameters in parallel (one a Fischer and Porter-Tube No. FP-2-27-G-10/80 Float No. 2" GNSVT 96—the other a Brooks-Tube No. R10M-25-3 Float No. 10LJ 238) and then through a 1" I. D. rubber hose to the mixing section. A six inch diameter aluminum cyclone separator was attached to the top of the storage drum so that the returning liquid would flow directly from the separator into the drum.

2. Gas System

The air supply was a 1" 100 psig dry air line. The air passed through a double reducing station, was heated by a heating tape, filtered through a Norgren 25 micron filter and metered through a Fischer and Porter rotameter (Float No. 1" GSVT 64-Tube No. B6-35-10/77). It passed through a 1" check valve and through a 1" I. D. rubber hose on to the mixing section.

3. Ammonia System

An anhydrous ammonia cylinder supplied ammonia which was regulated by an Olin Matheson valve No. 12-240 and metered through a F. and P. rotameter No. 448-209 into the air stream prior to the mixing section.

4. Pressure Drop Measurements

The pressure drop within the test section was measured by a Meriam 36" manometer filled with Meriam No. 3 Red Oil (S.G. = 2.95). Pressure taps were drilled and tapped for 1/8" N.P.T. fittings which were set flush with the inside of the test section. Clear Tygon tubing

was used for the tap lines so that air bubbles could be detected. A capacitance in the line was used to damp out oscillations in pressure. A U. S. Gauge (0 to 15psig) pressure gauge measured the pressure at one of the taps.

5. Electrical Conductivity Cells

The conductivity cells shown in Figs. 31 and 32 were used to measure the ammonia absorption. The 1/8" sampling taps were fitted with 1/8" Hoke valves which had both ends filled and redrilled to 1/32". One end of the Hoke valve was flush with the inside wall of the test section; the other end was flush with the middle section of the lucite conductivity cells as shown in the photograph (Fig. 32). The nickel electrodes were 1/4" x 1/8" x 1/16" with a gap of 1/8" between them. The sample passed through the Hoke valve into the conductivity cell, and on through another 1/8" Hoke valve and a Brooks Purge meter.

Figure 33 shows the 1000 cycle power source which applied a voltage across the cells and the amplifier which amplified and rectified the signals. The signal was sent to an L and N Speedomax-Type G multirange voltage recorder. There were zero, sensitivity, and range adjustments on the amplifier. The amplifier was zeroed with no input signal. The sensitivity and range adjustments were made only once so that a constant amplification resulted and calibration curves of recorder response versus concentration could be used for each cell. The cells were calibrated for recorder response as a function of concentration and temperature with known solutions of ammonia in distilled water. The output from the amplifier was in the range of 0 to 1 volt. A bucking voltage was used to buck off enough of this signal so that the remainder could be displayed on the 100 mv scale. Thus the response could be determined to ± 1 mv.

The ammonia concentration built up within the system during the run, doubling in approximately two minutes. However, due to the small concentrations, the equilibrium partial pressure above the liquid was always negligible. Distilled water was used for all ammonia

MU-34473

Fig. 31. Electrical conductivity cell and sampling system.

ZN-4400

Fig. 32. Photograph of conductivity cell.

experiments because the conductivity of tap water was comparable to that of the dilute ammonia concentrations measured.

The response of the cells to a step input was measured; the responses were identical for all three cells. The amount of sample between the electrodes was 0.1 cc. Due to an irregular flow pattern through the Hoke valve and some channeling within the cell itself, 2 cc. had to be passed through the cells to achieve 95 percent response to a step input. A number of runs were conducted varying the absorption time. They showed that $(NTU)_G$ was constant for times greater than 1-1/2 minutes. Thus operation for 2 minutes at 2 cc/min was enough to insure that the response represented concentrations existing at the sampling points.

6. Polarographic Oxygen Cells

The polarographic reduction of molecular oxygen at an electrode surface has been reported by many investigators.^{55,56} It was used here to measure the concentration of dissolved oxygen in water. Many electrode designs have been reported for various systems; the one used here follows that of Thomsen.⁵⁷

The design of the electrode system is shown in Figs. 34 through 37. The sampling line consisted of a 1/8" nipple filled with Scotchcast No. 2 epoxy resin with a 1/64" hole drilled through it. One end of the nipple was flush with the inside of the test section, the other was in the sampling cell system. The liquid flowed through the 1/64" hole into the semicircular passage in the sampling cell where it flowed past the 0.001" Teflon membrane of the electrode system and then through a Brooks Purge meter. The Bakelite electrode (Figs. 35 and 37) had a 1/16" x 1/32" disc platinum cathode and a 1' length of 0.02" diam. silver wire for the anode. The electrode was fit into an electrode holder and tightened down by a cap. The 0.001" Teflon membrane was held over the end of the electrode holder by an aluminum retaining ring. A 0.01 N KOH electrolyte filled the gap between the electrode and the electrode holder. The electrode and holder system was set into

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Fig. 34. Cross section of polarographic cell.

MU-34475

Fig. 35. Cross section of polarographic electrode.

Bakelite

MU-34476

Fig. 36. Cross section of electrode holder.

ZN-4401

Fig. 37. Photograph of electrode.

the sampling cell and tightened down so that the membrane formed an upper wall to the semicircular flow passage through the sampling cell. A 10K thermistor (Veco 41A3) was embedded in the electrode close to the platinum disc. It had a temperature coefficient of -4.1 percent per degree Centigrade which was equal and opposite to that of the uncompensated electrode system.⁵⁷

A voltage of -1.0 V. was applied across each cell. The current through the cell was measured by measuring the voltage drop across the 10K thermistor with a high impedance L and N Speedomax Type G voltage recorder. The circuitry is shown in Fig. 38.

The Teflon membrane being permeable to dissolved molecular oxygen permits the passage of oxygen to the platinum cathode where it is reduced to hydroxyl.

An Ag-Ag₂O reference electrode and KOH electrolyte were chosen because hydroxyl is the reaction product formed at the cathode.

a. Current-voltage curve. A typical current voltage curve is shown in Fig. 39. As the voltage is increased negatively from zero, more and more oxygen is reduced until at about -0.80 V. the oxygen concentration at the electrode has gone to zero and the diffusion of oxygen through the membrane limits the amount of oxygen reduced and therefore the current through the cell. This diffusion limited plateau persists until at even higher voltages water itself is reduced. Within the diffusion plateau the response should theoretically be proportional to oxygen concentration in the stream passing by the membrane and independent of voltage. It has been found that even though the plateau has a small slope, the height is proportional to concentration if the voltage is kept constant.^{56,58,59} The voltage was kept constant and the reading at steady state was assumed to be proportional to the oxygen concentration.

Bucking voltage 0 to 1 V

MU-34477

Fig. 38. Circuit diagram for polarographic electrodes.

MU-34478

Fig. 39. Current-voltage curve.

b. Temperature compensation. Thomsen found the temperature coefficient of his electrode to be +4 percent per degree Centigrade. The Veco 41A3 10K thermistor of temperature coefficient -4.1 percent per degree Centigrade provides temperature compensation. Figure 40 shows the degree of compensation affected by this method. Since the oxygen experiments were run isothermally, temperature compensation was not entirely necessary. However, it did enable the readings to be interpreted more easily.

c. Time constant. Steady state was achieved within a couple of minutes. This corresponds to a time constant in agreement with that calculated on the assumption that the entire resistance to diffusion is in the membrane.⁵⁷

d. Flow rate. Previous electrode systems have shown a dependence of diffusion current on the velocity of flow past the membrane.^{55,59} Such an effect was noted here and the rate of flow through the sampling cell was kept constant during a run.

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Fig. 40. Temperature compensated electrode: points are referred to left ordinate; line to right ordinate.

B. Regression Analysis

The least squares regression program utilized the SHARE MLR (minimum linear residuals) subroutine which determines $X(j)$ such that

$$\sum_{i=1}^m [R(i)]^2$$

is a minimum where

$$R(i) = Y(i) - \sum_{j=1}^n A(i,j)X(j), \quad (i = 1, 2, \dots, m), m \geq n, n > 1. \quad (103)$$

For a three constant case

$$Y_i = A(i,1)X(1) + A(i,2)X(2) + A(i,3)X(3) \quad (104)$$

To fit the data to the form

$$y_i = a x_i^b z_i^c \quad (105)$$

in order to determine the constants a , b and c logarithms may be taken of both sides of Eq. (105), giving

$$\log_{10} y_i = \log_{10} a + b \log_{10} x_i + c \log_{10} z_i. \quad (106)$$

If $A(i,1) = 1.0$, Eqs. (104) and (106) are in the same form.

C. Data

Table IX. Pressure Drop Data for Two Phase, Gas-Liquid Flow.

Nrun	number of run
W_o	mass flow rate of oil (lb/hr)
W_L	mass flow rate of water (lb/hr)
W_G	mass flow rate of air (lb/hr)
T	flow temperature (°F.)
P	average pressure (psia)
$(dP/dl)_{\text{exptl}}$	measured pressure gradient (psf/ft)
$(dP/dl)_{\text{calc}}$	pressure gradient calculated by correlation of the present study (psf/ft)
$2R_p$	inside diameter of tube or pipe (ft)
l	distance between pressure taps (ft)
f	Fanning friction factor
Re	Reynolds number (Equation (43))

Data of Martin -

$$2R_p = 0.0833 \quad l = 4.0$$

$$\text{Re} < 25,000 \quad f = 8.80 \times 10^{-3} (8,000/\text{Re})^{0.373}$$

$$\text{Re} \geq 25,000 \quad f = 4.60 \times 10^{-3} (110,000/\text{Re})^{0.156}$$

Air-Water Flow -

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>$(dP/dl)_{\text{exptl}}$</u>	<u>$(dP/dl)_{\text{calc}}$</u>
1	19000.	37.7	100.	23.2	149.	104.
2	19000.	35.7	100.	22.7	136.	102.
3	15120.	7.2	79.	17.8	52.	43.
4	15100.	19.4	79.	19.1	80.	57.
5	15120.	31.8	79.	20.3	89.	71.
6	15070.	40.2	79.	21.3	120.	78.
7	15060.	51.4	79.	22.8	138.	87.
8	18720.	41.1	78.	23.5	143.	105.

Table IX. (continued)

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>(dP/dℓ)</u> <u>exptl</u>	<u>(dP/dℓ)</u> <u>calc</u>
9	19000.	24.2	78.	22.3	114.	85.
10	19100.	17.5	78.	20.4	99.	79.
11	9250.	5.2	78.	15.7	25.	18.
12	9360.	16.4	78.	17.2	40.	26.
13	9370.	10.9	78.	16.7	34.	23.
14	9280.	22.5	78.	17.3	51.	30.
15	9300.	28.5	78.	17.9	60.	34.
16	5000.	8.2	75.	15.0	13.	8.
17	5000.	15.7	75.	15.4	23.	11.
18	5000.	24.2	75.	15.7	29.	13.
19	5000.	33.4	75.	16.1	35.	16.
20	5000.	36.0	75.	16.1	36.	17.
21	8950.	37.9	75.	17.6	64.	39.
22	9050.	55.8	75.	18.8	86.	49.
23	9050.	70.1	75.	19.2	95.	57.
24	15070.	68.8	75.	23.3	158.	105.
25	10250.	11.8	189.	17.4	69.	44.
26	10250.	19.7	191.	18.9	91.	56.
27	10250.	34.5	188.	20.6	110.	66.
28	10250.	36.6	190.	21.3	118.	68.
29	10250.	55.9	192.	22.9	138.	91.
30	15680.	18.7	190.	21.2	120.	80.
31	15680.	24.8	190.	22.9	142.	86.
32	15680.	46.9	190.	26.1	181.	112.
33	15630.	51.9	190.	27.1	192.	114.
34	11950.	10.0	190.	18.8	71.	44.
35	11950.	16.1	191.	20.0	92.	54.
36	11950.	24.8	191.	21.4	115.	64.
37	11850.	30.0	191.	22.1	130.	69.
38	13000.	10.6	190.	19.5	78.	48.
39	13100.	16.2	191.	20.6	99.	59.
40	13100.	22.7	191.	21.7	115.	68.
41	13100.	28.5	191.	22.6	130.	74.
42	14010.	9.0	190.	19.5	78.	50.
43	13950.	15.7	190.	20.9	102.	61.
44	14040.	20.7	191.	21.8	114.	70.
45	14040.	23.8	191.	22.5	129.	73.
46	14930.	8.4	190.	19.9	80.	52.
47	15050.	12.2	191.	20.8	98.	61.
48	15000.	17.7	190.	22.0	115.	68.
49	15070.	22.6	190.	22.9	125.	75.
50	16030.	7.8	189.	20.3	83.	54.
51	15980.	13.4	190.	21.7	104.	65.
52	15980.	18.2	190.	22.6	117.	73.
53	16050.	21.2	190.	23.3	132.	78.

Table IX. (continued)

$$\rho_{oil} = 52.7 \text{ lb/ft}^3 \text{ @ } 120^\circ \text{ F.}$$

$$\text{Air-Oil Flow - } \mu_{oil} = 24.0 \text{ lb/ft-hr @ } 120^\circ \text{ F.}$$

<u>Nrun</u>	<u>W_O</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	(dP/dl) <u>exptl</u>	(dP/dl) <u>calc</u>
317	12000.	8.4	120.	18.2	73.	64.
318	12000.	15.6	120.	19.1	90.	76.
319	11900.	24.5	120.	19.9	107.	89.
320	11800.	37.0	120.	21.0	126.	105.
321	14000.	12.3	120.	19.4	99.	87.
322	14000.	21.0	120.	20.4	116.	103.
323	13800.	28.8	120.	21.2	128.	114.
324	14000.	36.0	120.	22.0	141.	127.
325	15900.	9.5	122.	20.1	109.	96.
326	16100.	18.0	122.	21.0	131.	116.
327	15900.	24.8	122.	21.7	144.	127.
328	15800.	34.4	122.	22.6	159.	144.
329	17900.	10.3	122.	21.4	129.	115.
330	17800.	19.5	122.	22.4	151.	134.
331	17900.	25.9	122.	23.2	166.	149.
332	17900.	32.2	122.	24.0	179.	161.
333	20000.	11.3	122.	22.6	154.	136.
334	20000.	17.3	122.	23.9	171.	150.
335	20000.	25.0	122.	24.6	186.	167.
336	20000.	30.4	122.	25.4	201.	178.
338	12200.	7.4	118.	18.5	70.	64.
339	12000.	22.8	118.	19.1	90.	90.
340	12000.	24.9	118.	20.2	106.	91.
341	11800.	27.6	118.	20.5	110.	93.
344	14000.	7.2	118.	19.0	86.	77.
345	14000.	16.2	118.	20.0	106.	95.
346	14000.	24.4	118.	20.8	122.	110.
347	13900.	25.1	118.	20.9	122.	110.
350	16100.	7.2	118.	19.9	102.	94.
351	16000.	15.6	118.	20.7	123.	113.
352	16000.	20.8	118.	21.5	135.	123.
353	15900.	22.0	118.	21.6	136.	124.
356	18100.	5.4	118.	20.6	116.	107.
357	17800.	11.4	118.	21.4	133.	119.
358	18000.	16.4	118.	22.1	147.	132.
359	18000.	18.8	118.	22.5	152.	138.
362	20000.	6.6	118.	21.6	139.	128.
363	19800.	9.9	118.	22.6	149.	134.
364	20000.	13.6	118.	23.0	161.	145.

Table IX. (continued)

Data of James and Silberman -

$$2R_P = 0.206, \ell = 12.0, f = 4.24 \times 10^{-3} (34,800/\text{Re})^{0.0722}$$

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>(dP/dℓ)</u> <u>exptl</u>	<u>(dP/dℓ)</u> <u>calc</u>
600	122800.	150.5	77.	35.9	36.	30.
601	122800.	200.5	77.	35.6	40.	33.
602	122800.	258.0	77.	35.4	45.	36.
603	122800.	318.5	77.	35.1	51.	40.
604	163100.	94.4	77.	37.3	49.	44.
605	163100.	131.3	77.	37.0	55.	47.
606	163100.	173.5	77.	36.8	61.	51.

Data of Chisholm and Laird -

$$2R_P = 0.0886, \ell = 8.0, f = 6.00 \times 10^{-3} (29,500/\text{Re})^{0.246}$$

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>(dP/dℓ)</u> <u>exptl</u>	<u>(dP/dℓ)</u> <u>calc</u>
700	9040.	4.6	61.	17.5	16.	13.
701	9040.	8.4	61.	17.5	18.	15.
702	9040.	11.2	61.	17.5	21.	16.
703	9040.	17.3	61.	17.7	25.	19.
704	12650.	4.7	61.	17.9	28.	22.
705	12650.	8.2	61.	18.4	32.	24.
706	12650.	11.9	61.	18.4	34.	27.
708	12650.	17.3	61.	19.2	38.	30.
709	12650.	27.8	68.	20.9	49.	35.
710	12650.	49.0	68.	22.2	59.	45.

Data of Johnson and Abou-Sabe -

$$2R_P = 0.0725, \ell = 15.7, f = 0.00140 + 0.125/\text{Re}^{0.32}$$

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>(dP/dℓ)</u> <u>exptl</u>	<u>(dP/dℓ)</u> <u>calc</u>
801	15000.	7.6	69.	26.7	92.	76.
802	15000.	14.8	67.	30.7	106.	85.
803	15000.	27.0	64.	35.7	128.	98.
804	15000.	67.5	73.	41.7	161.	134.
805	15000.	105.0	79.	48.7	188.	154.
806	15000.	177.0	84.	61.7	238.	180.
807	9000.	14.4	82.	23.7	61.	41.
808	9000.	24.2	80.	26.7	80.	48.
809	9000.	57.1	78.	30.7	108.	70.
810	9000.	111.0	76.	37.7	152.	94.
811	9000.	189.7	74.	47.7	169.	114.

Table IX. (continued)

Data of present work.

$$2R_P = 0.0833 \quad l = 3.0^* \quad f = 4.00 \times 10^{-3} (130,000/Re)^{0.0702}$$

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>(dP/d l) exptl</u>	<u>(dP/d l) calc</u>
500	15300.	46.2	75.0	16.7	110.	100.
501	15300.	46.2	75.0	17.9	99.	92.
502	17650.	45.0	75.0	19.2	127.	108.
503	17650.	45.0	75.0	17.2	142.	123.
504	21150.	43.3	75.0	21.4	187.	130.
505	21150.	43.3	75.0	18.4	205.	155.
508	17850.	109.0	75.0	22.5	212.	212.
511	19450.	106.0	75.0	24.2	255.	214.
512	11500.	72.0	75.0	16.8	88.	89.
513	12300.	71.5	75.0	17.4	100.	95.
514	15400.	70.0	75.0	18.3	135.	134.
515	16900.	69.3	79.0	19.0	160.	151.
516	18150.	69.0	79.0	19.7	188.	163.
517	19400.	67.0	79.0	20.4	218.	171.
518	20650.	67.5	80.0	21.0	238.	187.
523	18700.	107.0	80.0	21.4	267.	266.
524	19750.	103.0	80.0	21.9	292.	270.

Runs having $(dP/d l)_{\text{accel}} > (dP/d l)_{\text{frict}}$

These runs lie outside line 4 on Figure 12.

506	15300.	114.0	75.0	19.0	142.	239.
507	15300.	114.0	75.0	20.7	156.	192.
509	17850.	109.0	75.0	19.4	233.	312.
510	19450.	106.0	75.0	20.2	290.	329.
519	11250.	115.0	80.0	17.7	101.	140.
520	12300.	117.5	81.0	17.9	123.	174.
521	14950.	112.0	81.0	19.0	159.	222.
522	16900.	109.0	81.0	20.4	222.	243.
525	10250.	181.5	81.0	18.0	110.	266.
526	12300.	169.0	82.0	18.8	153.	333.
527	16900.	159.0	82.0	21.3	273.	463.
528	18700.	153.5	82.0	23.1	337.	414.
529	18150.	192.0	83.0	23.7	363.	606.
530	12300.	224.0	83.0	19.4	186.	878.
531	11900.	268.0	83.0	19.7	191.	3931.

*
Runs 500, 503, 505, 506, 509, and 510 were measured over the last foot of the 3 ft. section

Runs 501, 502, 504, 507, 508 and 511 were measured over the first one foot of the 3 ft. section.

Table IX. (continued)

Data of present work

$$2R_P = 0.0833 \quad l = 1.0 \quad f = 7.60 \times 10^{-3} (100,000/Re)^{0.21}$$

<u>Nrun</u>	<u>W_L</u>	<u>W_G</u>	<u>T</u>	<u>P</u>	<u>(dP/dl) exptl</u>	<u>(dP/dl) calc</u>
675	11300.	30.2	78.0	16.7	76.	83.
676	11300.	40.7	78.0	16.8	81.	100.
678	14350.	40.8	78.0	18.7	127.	133.
679	16400.	40.0	78.0	19.6	165.	155.
680	18450.	40.0	78.0	20.6	203.	180.
681	18850.	27.8	78.0	20.2	182.	152.
682	16400.	28.3	78.0	18.9	145.	129.
683	14350.	28.6	78.0	18.1	113.	109.
684	12300.	29.0	78.0	17.1	85.	90.
685	11300.	28.8	78.0	16.7	74.	80.
686	11300.	16.6	78.0	16.3	66.	60.
687	14350.	15.9	78.0	17.3	98.	83.
688	16400.	16.2	78.0	18.2	123.	101.
689	18450.	15.4	78.0	18.8	149.	118.
690	12300.	39.2	78.0	17.6	92.	107.
691	11300.	57.0	78.0	17.7	93.	124.
692	14350.	57.2	78.0	19.4	146.	166.
693	16400.	55.5	78.0	20.7	187.	187.
694	18450.	54.0	78.0	21.7	233.	212.
695	11300.	69.0	78.0	17.9	99.	145.
696	14350.	66.5	78.0	19.8	163.	184.
697	16400.	65.5	78.0	21.2	204.	209.
698	18450.	64.0	78.0	22.6	256.	232.
 <i>l</i> = 2.0						
600	12300.	32.0	88.0	17.0	62.	66.
601	14350.	42.7	72.0	18.3	98.	96.
602	12100.	35.4	86.0	17.2	63.	68.
603	11250.	43.5	78.0	17.1	65.	71.
604	9400.	45.3	82.0	16.3	48.	58.

Table X. Ammonia Absorption Data

Run	W_L (lb/hr)	W_G (lb/hr)	NH_3 $\left(\frac{\text{lb-moles}}{\text{hr}}\right)$	l (ft)	T_L (°C)	T_G (°C)	P_1 (psia)	ΔP (psia)	Conc. _g NH_3 (m/l)	Conc. ₁ NH_3 (m/l)	Conc. ₂ NH_3 (m/l)	(NTU) _{OG} corrected to 25° C
1	11,300.	41.2	0.0396	1.0	28.0	24.5	16.9	0.50	0.00017	0.00251	0.00312	0.86
2	11,300.	54.2	0.0436	1.0	27.0	26.0	17.25	0.55	0.00060	0.00265	0.00371	0.97
3	11,300.	65.8	0.0588	1.0	24.5	26.0	17.80	0.60	0.00082	0.00395	0.00538	1.35
4	11,300.	28.8	0.0363	1.0	23.8	24.0	16.45	0.45	0.00057	0.00258	0.00299	0.49
5	14,350.	28.7	0.0394	1.0	25.5	24.0	17.90	0.70	0.00071	0.00292	0.00310	0.56
6	14,350.	44.0	0.0398	1.0	27.2	27.5	18.5	0.80	0.00035	0.00255	0.00285	0.93
7	14,350.	55.3	0.0456	1.0	26.0	27.5	19.1	0.90	0.00070	0.00296	0.00349	1.02
8	14,350.	66.0	0.0569	1.0	24.8	26.0	19.7	1.00	0.00163	0.00482	0.00532	1.35
9	16,400.	31.3	0.0354	1.0	26.1	25.0	19.1	0.90	0.00650	0.00800	0.00825	0.60
10	16,400.	43.0	0.0461	1.0	21.4	23.0	19.8	1.00	0.00112	0.00335	0.00365	0.96
11	16,400.	57.6	0.0530	1.0	23.9	24.5	20.9	1.20	0.00590	0.00850	0.00887	1.14
12	16,400.	65.0	0.1080	1.0	25.8	26.3	21.15	1.25	0.00165	0.00700	0.00775	1.54
13	11,300.	40.0	0.0415	0.5	22.9	22.5	16.95	0.25	0.00175	0.00424	0.00467	0.48
14	14,350.	42.7	0.0493	0.5	24.8	25.0	18.60	0.40	0.00492	0.00778	0.00797	0.55
15	11,300.	54.4	0.0475	0.5	26.2	25.0	17.5	0.30	0.00067	0.00358	0.00413	0.635
16	14,350.	30.2	0.0385	0.5	26.6	24.8	18.15	0.35	0.00347	0.00555	0.00570	0.37
17	11,300.	63.2	0.0485	0.5	24.2	25.3	17.8	0.30	0.00083	0.00355	0.00430	0.68
18	11,300.	63.2	0.0485	0.5	28.4	28.0	17.75	0.30	0.00450	0.00750	0.00805	0.67
19	11,300.	63.2	0.0485	0.5	17.7	20.0	17.70	0.30	0.00145	0.00403	0.00503	0.675

Table XI. Oxygen absorption data

Run No.	W_L (lb/hr)	W_G (lb/hr)	l (ft)	T °C	P_1 (psia)	ΔP (psia)	c_1 (rel.)	c_2 (rel.)	$(NTU)_{OL}^c$
1	14,750.	37.3	1.0	30.0	18.90	0.40	1.04	1.21	0.95 ^a
2	15,370.	31.6	1.0	31.0	19.20	0.45	1.09	1.19	0.44 ⁵
3	17,600.	32.5	1.0	31.5	20.2	0.60	1.045	1.225	0.61
4	14,750.	44.0	1.0	32.0	19.0	0.40	1.045	1.18	0.54
6	11,250.	74.5	1.0	25.0	18.7	0.35	1.10	1.195	0.79
7	12,100.	97.0	1.0	28.5	19.95	0.45	1.075	1.31	1.58
8	11,100.	69.0	1.0	27.5	18.20	0.30	1.09	1.185	0.85
9	11,700.	42.7	1.0	28.5	17.6	0.25	1.09	1.135	0.43
10	11,450.	40.5	1.0	29.0	17.2	0.25	1.05	1.095	0.38
11	14,150.	41.0	1.0	28.0	18.8	0.40	1.06	1.18	0.67
12	15,750.	30.2	1.0	29.0	19.85	0.50	1.03	1.265	1.09 ^a
13	12,300.	32.0	2.0	31.5	17.45	0.85	1.09	1.13	0.87 ^b
14	14,350.	42.5	2.0	22.5	18.95	1.35	1.075	1.19	1.58 ^b
15	12,100.	35.4	2.0	30.0	17.7	0.90	1.08	1.14	0.97 ^b
16	11,250.	43.5	2.0	26.0	17.5	0.90	1.045	1.115	0.97 ^b
18	9,400.	45.3	2.0	28.0	16.7	0.65	1.045	1.08	0.63 ^b
19	14,350.	42.5	1.0	28.0	18.95	0.75	1.075	1.185	0.66
20	11,000.	61.0	1.0	33.0	17.70	0.55	1.055	1.14	0.60
21	14,350.	28.6	1.0	34.0	18.15	0.70	1.05	1.135	0.40
22	14,350.	40.8	1.0	28.0	18.95	0.80	1.05	1.195	0.89 ^a
23	14,350.	56.3	1.0	30.0	19.7	0.90	1.055	1.24	0.91
24	16,400.	28.2	1.0	27.5	19.20	0.95	1.06	1.17	0.50

^a rejected when compared to similar runs^b large uncertainty because of close approach to equilibrium^c corrected to 25°C

Table XI. (continued)

Run No.	W_L (lb/hr)	W_G (lb/hr)	l (ft)	T (°C)	P_1 (psia)	ΔP (psia)	c_1 (rel.)	c_2 (rel.)	$(NTU)_{OL}^c$
25	14,350.	66.5	1.0	34.0	20.2	1.05	1.075	1.29	0.99
26	16,400.	41.7	1.0	24.5	20.15	1.10	1.06	1.265	1.27 ^a
27	16,400.	55.6	1.0	23.5	21.1	1.20	1.08	1.33	1.45 ^a
28	12,300.	55.0	1.0	25.0	18.15	0.70	1.04	1.18	1.27 ^a
29	12,300.	66.5	1.0	31.0	18.8	0.75	1.04	1.21	1.04
30	15,350.	65.5	1.0	33.0	20.95	1.20	1.06	1.33	1.09
31	12,300.	32.0	1.0	27.5	17.1	0.30	1.055	1.09	0.31
32	11,250.	32.0	1.0	30.0	16.65	0.40	1.04	1.07	0.28
33	11,250.	57.0	1.0	29.0	17.1	0.60	1.055	1.125	0.54
34	12,300.	56.0	1.0	32.0	17.7	0.70	1.055	1.17	0.65
35	12,300.	66.0	1.0	34.0	18.0	0.80	1.045	1.20	0.81
36	14,750.	42.0	1.0	24.0	18.9	0.80	1.065	1.165	0.66
37	14,750.	42.0	1.0	29.5	18.9	0.80	1.06	1.19	0.73
38	14,750.	42.0	1.0	34.0	18.9	0.80	1.04	1.195	0.725
39	14,750.	42.0	1.0	19.0	18.9	0.80	1.045	1.145	0.71
40	14,750.	42.0	0.5	23.0	18.5	0.40	1.14	1.18	0.43
41	14,750.	42.0	0.5	26.0	18.5	0.40	1.145	1.19	0.46 ^b
42	11,300.	42.0	0.5	29.0	16.9	0.25	1.11	1.125	0.29 ^b
43	14,750.	56.0	0.5	31.0	19.0	0.40	1.115	1.21	0.55
44	15,400.	42.0	0.5	25.0	18.8	0.40	1.14	1.20	0.55
45	16,400.	42.0	0.5	29.0	19.5	0.50	1.12	1.21	0.46

^a rejected when compared to runs at similar flow rates^b large uncertainty because of close approach to equilibrium^c corrected to 25° C.

D. Sample Calculations

1. Calculation of $(NTU)_{OL}$ for run No. 19:

$$\begin{aligned} W_L &= 14,350. \text{ lb/hr} & \Delta P &= 0.75 \text{ psi} \\ W_G &= 42.5 \text{ lb/hr} & c_1 &= 1.075 \text{ (rel.)} \\ T &= 28^\circ\text{C} & c_2 &= 1.185 \text{ (rel.)} \\ P_1 &= 18.95 \text{ psia} \end{aligned}$$

c_1 and c_2 are relative concentrations defined as

$$c(\text{rel.}) = c/c_{\text{ebase}}$$

where c_{ebase} is the oxygen concentration in equilibrium with air at the pressure $(14.7 - p_w)$ psia where p_w is the vapor pressure of water at T of the run. For run 19, $p_w = 0.55$ psi. Therefore c_{ebase} is given by

$$c_{\text{ebase}} = \frac{0.21(14.7 - 0.55)}{H}$$

c_{e1} is given by

$$c_{e1}(\text{rel.}) = (P_1 - p_{w1})/14.15$$

and

$$c_{e2}(\text{rel.}) = (P_2 - p_{w2})/14.15 .$$

Since $P_1 = 18.95$, and $\Delta P = 0.75$

$$P_2 = P_1 - \Delta P = 18.20 .$$

We estimate p_{w1} as 0.35 and p_{w2} as 0.50 so that

$$c_{e1}(\text{rel.}) = (18.95 - 0.35)/14.15 = 1.315$$

$$c_{e2}(\text{rel.}) = (18.20 - 0.50)/14.15 = 1.25$$

$$\begin{aligned}
 (NTU)_{OL}^A &= \frac{c_2 - c_1}{(c_{e1} - c_1) - (c_{e2} - c_2)} \ln \frac{(c_{e1} - c_1)}{(c_{e2} - c_2)} \\
 &= \frac{1.185 - 1.075}{(1.315 - 1.075) - (1.25 - 1.185)} \ln \frac{(1.315 - 1.075)}{(1.25 - 1.185)} \\
 &= 0.82
 \end{aligned}$$

$$\begin{aligned}
 (NTU)_{OL}^C &= \ln \frac{(c_{e1} - c_1)}{(c_{e1} - c_2)} \\
 &= \ln \frac{(1.315 - 1.075)}{(1.315 - 1.185)} \\
 &= 0.61
 \end{aligned}$$

$$\begin{aligned}
 (NTU)_{OL} &= (NTU)_{OL}^A - 0.35((NTU)_{OL}^A - (NTU)_{OL}^C) \\
 &= 0.82 - 0.35(0.82 - 0.61) \\
 &= 0.75
 \end{aligned}$$

$$(NTU)_{OL} = 0.75e^{0.042(25-T)}$$

$$\begin{aligned}
 \text{corrected} &= 0.66 \\
 \text{to } 25^\circ\text{C}
 \end{aligned}$$

2. Calculation of $(NTU)_{OG}$ for run No. 17:

$W_L = 11,300. \text{ lb/hr}$	$\text{Conc}_s = 0.00083 \text{ m/l}$
$W_G = 63.2 \text{ lb/hr}$	$\text{Conc}_1 = 0.00355 \text{ m/l}$
$\text{NH}_3 = 0.0485 \text{ lb-moles/hr}$	$\text{Conc}_2 = 0.00430 \text{ m/l}$
$T_L = 24.2^\circ\text{C}$	
$P_1 = 17.8 \text{ psia}$	
$\Delta P = 0.30 \text{ psi}$	

The material balances are

$$W_{LM}(x_1 - x_s) = W_{GM}(y_s - y_1)$$

and

$$W_{LM}(x_2 - x_1) = W_{GM}(y_1 - y_2)$$

where

$$W_{LM} = 11,300/18 = 627 \text{ lb-moles/hr}$$

$$W_{GM} = 63.2/29 + 0.0485 = 2.25 \text{ lb-moles/hr}$$

W_{GM} changes slightly as water is vaporized and ammonia is absorbed, however, these effects approximately cancel each other out.

The mole fraction x is given by

$$x = \frac{\text{conc(m/l)}(18)}{1000}$$

so that

$$x_s = 1.50 \times 10^{-5}$$

$$x_1 = 6.40 \times 10^{-5}$$

$$x_2 = 7.78 \times 10^{-5}$$

and finally

$$y_s = 0.0485/2.25 = 0.0215.$$

Substituting into the material balances, we arrive at

$$y_1 = 0.0078$$

$$y_2 = 0.00395$$

Since the derivation of Eq. (22) assumes constant P , we take

$$p_1 = y_1 P_{\text{ave}} = (0.0078)(17.65) = 0.138 \text{ psi}$$

$$p_2 = y_2 P_{\text{ave}} = (0.00395)(17.65) = 0.07 \text{ psi .}$$

Finally

$$\begin{aligned} (\text{NTU})_{\text{OG}} &= \ln p_1/p_2 = \ln 0.138/0.07 \\ &= 0.68 \end{aligned}$$

and

$$\begin{aligned} (\text{NTU})_{\text{OG}} &= 0.68 e^{-0.011(25-T)} \\ \text{corrected} &= 0.68 \\ \text{to } 25^{\circ}\text{C} \end{aligned}$$

E. Nomenclature

A	cross-section area of tube, ft^2
a	interfacial area per unit volume, ft^{-1}
a.d.	average deviation, dimensionless
B	constant in Blasius equation, dimensionless
c	solute concentration in liquid, lb-moles/vol
c_e	solute concentration in liquid in equilibrium with gas, lb-moles/vol
D	diffusivity, ft^2/hr
d	bubble diameter, ft
ΔP	pressure difference between taps, psf
$dP/d\ell$	pressure gradient, psf/ft
err	$\frac{(\frac{dP}{d\ell})_{\text{calc}} - (\frac{dP}{d\ell})_{\text{exptl}}}{(\frac{dP}{d\ell})_{\text{exptl}}} (100), \text{ dimensionless}$
f	Fanning friction factor, dimensionless
Fr	Froude Number, dimensionless
G	superficial mass velocity of air, lb/hr-ft^2
g	acceleration of gravity, ft/sec^2
g_c	gravitational constant, lbm-ft/lbf-sec^2
K	ratio of <u>in situ</u> void fraction to void fraction in feed, dimensionless
k_c	mass transfer coefficient equal to k_{RT} , ft/hr
K_G	overall coefficient, $\text{lb moles/hr-ft}^2\text{-atm}$
K_G^a	overall coefficient, $\text{lb moles/hr-ft}^2\text{-atm}$
k_G	gas film coefficient, $\text{lb moles/hr-ft}^2\text{-atm}$
k_G^a	gas film coefficient on a volume basis, $\text{lb moles/hr-ft}^3\text{-atm}$
K_L	overall coefficient, $\text{lb moles/hr-ft}^2\text{-unit } \Delta c$
K_L^a	overall coefficient on a volume basis, $\text{lb moles/hr-ft}^3\text{-unit } \Delta c$

k_L	liquid film coefficient, lb moles/hr-ft ² -unit Δc
k_L^a	liquid film coefficient on a volume basis, lb moles/hr-ft ³ -unit Δc
L	superficial mass velocity of liquid, lb/hr-ft ²
ℓ	length of test section, ft
$(LTU)_L$ or G	length of an individual phase transfer unit, ft
$(LUT)_{OL}$ or OG	length of an overall transfer unit, ft
m	dy_e/dx , slope of equilibrium curve, dimensionless
n	exponent in Blasius equation, dimensionless
N_A	diffusion rate, lb moles/hr-ft ²
$(NTU)_L$ or G	number of individual phase transfer units, dimensionless
$(NTU)_{OL}$ or OG	number of overall transfer units, dimensionless
P	total pressure, atm
p	partial pressure of solute, atm
p_e	partial pressure of solute in equilibrium with liquid, atm
p_w	vapor pressure of water, atm
R_P	radius of tube, ft
R	gas constant, ft ³ -atm/lb moles-°R
Re	Reynolds Number, dimensionless
s	molality of ammonia, moles/liter
$s.d.$	standard deviation, dimensionless
Sc	Schmidt Number, dimensionless
Sh	Sherwood Number, dimensionless
T	temperature, °C
t	time, sec
U	velocity, ft/sec
W	mass flow rate. lb/hr
x	mole fraction of solute in liquid, dimensionless

x_e	mole fraction of solute in liquid in equilibrium with gas, dimensionless
X_v	weight fraction of vapor, dimensionless
Y	saturation defined by Eq. (60), dimensionless
y	mole fraction of solute in gas, dimensionless
y_e	mole fraction of solute in gas in equilibrium with liquid, dimensionless
Z	Hughmark holdup parameter, dimensionless
z	mean radial separation between bubbles, in.

Greek Letters

α	<u>in situ</u> void fraction, dimensionless
β	temperature coefficient, $^{\circ}\text{C}^{-1}$
δ	difference in concentration defined in Eq. (24), dimensionless
ϵ	energy dissipation per unit mass and time, ft^2/sec^3
η	local scale of turbulence, ft
Λ	density parameter, dimensionless
λ	linear concentration defined in Eq. (96), dimensionless
μ	viscosity, lb/hr-ft
ν	kinematic viscosity, centistokes
ρ	density, lb/ft^3
σ	surface tension, dynes/cm
ϕ	void fraction in feed, dimensionless
ψ	viscosity and surface tension parameter, dimensionless

Subscripts

accel	acceleration
ave	average
calc	calculated
ds	mean diameter surface
e	equilibrium
exptl	experimental

fric	frictional
G	gas
GS	superficial gas
hom	homogeneous
L	liquid
LS	superficial liquid
M	on a molar basis
m	two phase mixture
s	start of test section
sv	mean surface volume
TP	two phase mixture averaged over cross section
v	vapor
1	start of measuring section
2	end of measuring section
50	exceeded by 50 percent
95	exceeded by 5 percent

Superscripts

A	defined by Eq. (19)
C	defined by Eq. (24)
*	intermediate

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