

The Behavior of Immiscible Liquids in Concurrent Flow Through Packed Beds

ROBERT G. RIGG and STUART W. CHURCHILL

The University of Michigan, Ann Arbor, Michigan

In countercurrent flow of two fluids through a tube or packed bed the throughput is severely limited by the necessity of using density difference as a driving force for one of the fluids. In concurrent flow this limitation is avoided. A preceding investigation of liquid-liquid extraction in concurrent flow through a packed bed (1) indicated that the mass transfer coefficients for both phases were very high and increased indefinitely with throughput. This observation and the absence of any prior work suggested a study of the fluid mechanics of such systems.

The most important information for design purposes is the pressure gradient. A knowledge of the phase holdup and the drop size distribution is also important for an understanding of the momentum and mass transfer processes within the bed. These three dependent variables were accordingly measured as a function of the flow rates of the two phases. Measurements were made in upflow only, with two different liquid pairs plus a surfactant in order to give a relatively wide range of density, viscosity, and interfacial tension, and with glass spheres of three different sizes. The effects of column diameter, packing shape and composition, inlet arrangement, and direction of flow were not investigated in this initial study.

Complete details of the investigation and a bibliography of generally related work are given in reference 2.

EXPERIMENTAL APPARATUS

A schematic diagram of the experimental apparatus is shown in Figure 1. The measurements were carried out in a flanged, 82-in. long, 4-in. I.D. transparent Lucite tube filled with glass spheres of uniform size. This tube was mounted vertically between two 4 in., flanged, quick-closing valves. Retaining screens were placed inside the valves so that the valves could be filled with packing right up to the valve disk. The valves were closed rapidly and simultaneously by a falling weight connected by steel cables through pulleys to the valve handles.

Pressure taps consisting of $\frac{1}{8}$ -in. pipe and $\frac{3}{16}$ -in. copper tubing were located just below the lower valve disk and at four points along the column as indicated. Crossed wires were placed in the pipe connectors to prevent the packing from blocking the taps. Pressure measurements were made with a dual, well type of manometer with a 40-in. range. One tube was filled with mercury and the other with a manometer fluid, immiscible with water and having a specific gravity of 1.750 at 20°C. relative to water at 4°C. A manifold enabled either tube to be connected across any two pressure taps. Prior to pressure measurements the manometer leads were flushed with the water phase.

The two liquids were pumped separately from 200-gal. storage tanks through 8 and/or 20 gal./min. rotameters, glass wool filters, and check valves into two arms of a 2-in. cross.

The mixture flowed out a third arm of the cross into the lower quick-closing valve. The temperature of the mixture was measured in the cross with an immersion type of copper-constantan thermocouple in a $\frac{1}{4}$ -in. well. The effluent from the upper quick-closing valve passed through a $1\frac{1}{2}$ -in. pipe to the side of a 140-gal. separating tank. The separated liquids flowed by gravity to their respective storage tanks. The liquid interface was maintained approximately at the midpoint by controlling the drainage rate of the heavier liquid.

To avoid liquid hammer a bypass was provided from the fourth arm of the inlet cross through a check valve to a surge tank. Before a run the surge tank was filled with air to a pressure 10 lb./sq.in. greater than the expected pressure at the cross. When the quick-closing valve was actuated, the check valve was forced open and the liquid mixture was automatically diverted to the surge tank. The drain valve on the surge tank was then opened and the pumps turned off. A relief valve set at 100 lb./sq.in. was provided to protect the surge tank.

Packing

The packing consisted of uniformly sized glass spheres. The characteristic diameter of the spheres was determined by two

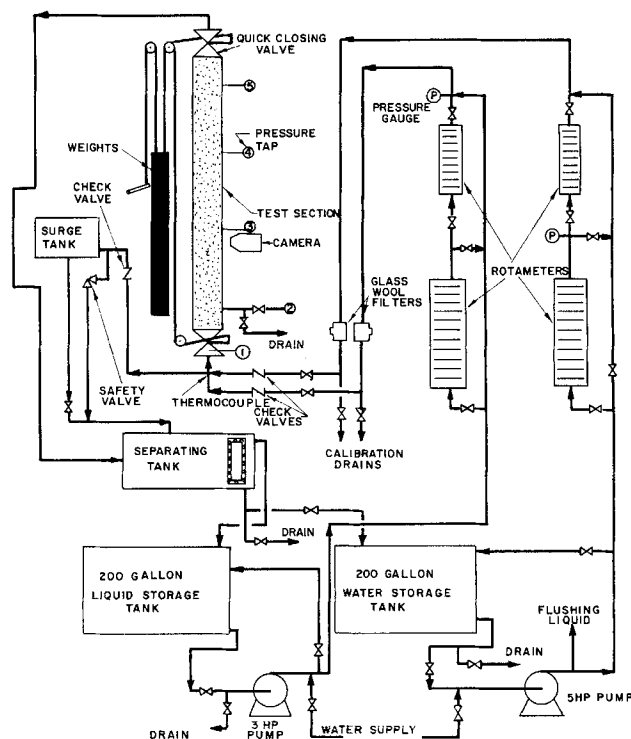


Fig. 1. Schematic diagram of experimental apparatus.

Robert G. Rigg is with the American Oil Company, Whiting, Indiana.

TABLE I. PROPERTIES OF PACKING

Diameter, in.	Glass density, g./cu. in.	Void volume (direct weighing)
0.501	40.57	0.400
0.340	40.89	0.383
		0.382*
0.164	47.98	0.337

* Obtained after column was repacked for water-isooctane tests.

methods. Fifty spheres of each size were selected at random, and the diameter of each sphere was measured five times in random directions with a micrometer. An arithmetic mean diameter was computed from these measurements. Also the total volume of the fifty spheres was determined by water displacement, and a mean diameter was computed with spherical shape assumed. In all cases the two determinations of the mean diameter agreed within 0.002 in. The glass density was computed by dividing the weight of the fifty spheres by their volume.

In order to obtain a reproducible packing arrangement the column was filled by settling the spheres through water while tapping the side of the column. The displaced water was collected and the volume determined. The porosity of the packed bed was then computed from the known dimensions of the test section. The porosity was also computed by weighing the packing. The maximum difference in the two determinations was 0.008. The characteristics of the packings are summarized in Table 1. The smallest spheres came from a different source and had a much greater density. The three packings are hereafter designated only by their nominal diameters of 0.164, 0.340, and 0.501 in.

Liquids

The liquid pairs which were utilized were mutually saturated water-isobutanol and water-isooctane. Commercial grade isobutanol and tap water were circulated through the system for 4 hr. to saturate both phases. The water-rich phase was then acidified to 0.0001 N hydrochloric acid to prevent the formation of stable emulsions. This small concentration of acid was not found to affect the physical properties of either phase significantly. The isooctane was pure grade and contained a minimum of 99 mole % 2,2,4-trimethyl pentane. A separate set of runs was made after the addition of 0.009 vol. % Alkaterge C, an organic-soluble surfactant, to the isooctane, thus reducing the interfacial tension significantly while maintaining the same densities and viscosities.

The relevant properties of the liquids are given in Table 2. The properties which were not available in the literature were measured, and the available values were checked, with good agreement in all cases. Viscosity was measured with viscometer and interfacial tension with a ring type of tensiometer.

EXPERIMENTAL MEASUREMENTS

The drop-size distribution of the dispersed phase was determined from flash photographs taken through the wall of the test section with a camera with a 50-mm. lens, an adjustable focus, and stops from $f/2.8$ to $f/22$. The resulting magnification was 2.94X. A ground glass focusing plate mounted in

the film holder enabled the droplet images to be focused on the film without trial and error. 4 in. \times 5 in. fine-grained, orthochromatic film and a high intensity flash tube were used. The camera was positioned approximately 24 in. above the inlet to the packed column. Transmitted light, which results in less distortion, was used with the 0.501-in. spheres, but the use of 45 deg. reflected light was necessary with the smaller spheres because of reduced transmission. At least two photographs were taken for each condition.

Drop sizes were measured directly from the negatives with a comparator which produced a 10X enlarged image on a ground glass screen thus producing an overall magnification of 29.4X. Droplet diameters were measured only in the direction of the axis of the test section because the wall of the test section produced some distortion perpendicular to the axis. Two triangular templates, one graduated in tenths of an inch to 1.5 in. and the other graduated in hundredths of an inch to 0.5 in., were used. All droplets in reasonable focus were measured, the number counted varying from 11 to 283.

Phase holdup was determined as follows. After the quick-closing valves were actuated, the phases were allowed to separate. If a clearly defined interface formed, the distance from the lower flange was measured. If a clear interface was not formed, as sometimes occurred with the smallest packing, the dense phase was allowed to drain slowly through a sample tap located at pressure tap No. 2 and the volume was measured. In both methods the phase holdup was then computed from geometrical considerations. The holdup of liquid at the points of contact of the packing amounted to a maximum of 2.65% of the void volume (with the smallest packing). Therefore it is concluded that capillary forces were not very significant and that the indicated phase holdups may be in error to the extent of 2.65%.

In the runs used to measure the transient development of the phase holdup, flow of one phase was first fully established and then flow of the second phase. After a short time the quick-closing valves were actuated and the phase holdup measured. This procedure was repeated for a series of increasing lengths of time.

The upper limit of flow rates was imposed by the characteristics of the pumps, the lower limit by the sensitivity of the manometers and rotameters. Approximately forty runs were carried out for each liquid pair and packing to cover the range of flows. Approximately ten runs were repeated to test reproducibility. Reproducibility was not characterized statistically, but the repeated data are included in the plots and correlations.

The pressure gradient over each of the four sections of the packed bed was computed from the manometer readings. The pressure gradient over the inlet section was in all cases the least. The pressure gradients over the other three sections were essentially equal, and the average of the three was used for correlation.

The raw data were processed, and all correlations were developed on the IBM-7090 at the University of Michigan Computing Center.

EXPERIMENTAL RESULTS

Single-Phase Flow

Data were obtained for single-phase flow to test the reliability of the experimental technique and to obtain a

TABLE 2. PHYSICAL PROPERTIES OF LIQUIDS

Liquid	Composition	Density at 75°F. (g./cc.)	Viscosity at 75°F. (centipoise)	Interfacial tension against saturated water solution at 75°F. (dynes/cm.)
Water	—	0.998 (5)	0.915 (7)	—
Water saturated with isobutanol	8.4 wt. % isobutanol (3)	0.987 (6)	1.30	—
Isobutanol saturated with water	83.5 wt. % isobutanol (3)	0.832 (6)	3.10	2.1 (8)
Isooctane	99 mole % min. 2,2,4-trimethyl pentane (4)	0.692	0.478	49.5 (9)
Isooctane with Alkaterge C	0.009 vol. % Alkaterge C	0.692 (5)	0.478	16.0

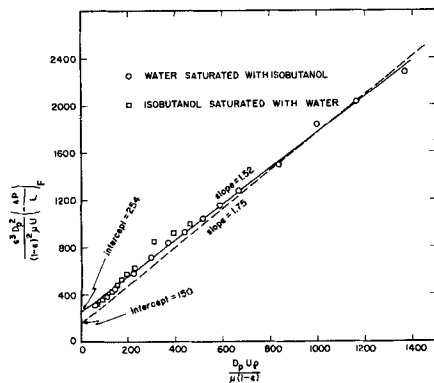


Fig. 2. Ergun type of correlation of single-phase pressure gradients, saturated isobutanol and water with 0.340-in. spheres.

single-phase correlation as a starting point for a two-phase correlation. Of the many correlations that have been proposed for the pressure gradient due to friction in single-phase flow through packed beds, the most theoretically suitable appears to be that of Ergun and Orning (10). On the basis of a simple physical model they derived the following equation:

$$\frac{\epsilon^3 D_p^2}{(1-\epsilon)^2 \mu U} \left(-\frac{\Delta P}{L} \right)_F = k_1 + k_2 \left(\frac{D_p U \rho}{\mu(1-\epsilon)} \right) \quad (1)$$

Ergun (11) determined values of 150 for k_1 and 1.75 for k_2 from data for air flowing through beds of spheres.

The coefficients k_1 and k_2 were determined by least squares from the single-phase data obtained in this investigation after subtracting the static pressure gradient from the measured gradient to obtain the gradient due to friction. As illustrated in Figure 2, the data for a single packing correlate very well in the linear form suggested by Equation (1), indicating that k_1 and k_2 are not functions of velocity or fluid properties. However, as indicated in Table 3 as well as Figure 2, the values of k_1 and k_2 differ from packing to packing and from the values proposed by Ergun (11). The wide variation in k_1 is probably not significant since the data herein were generally for relatively large $D_p U \rho / \mu$.

All of the single phase data are plotted in Figure 3 in logarithmic coordinates. The data are consistently lower than predicted by Ergun's coefficients at large $D_p U \rho / \mu$. The deviations are greatest for the 0.501-in. spheres and may be related to the relatively large ratio of the particle diameter to the column diameter. However an examination of the data from which Ergun derived the values of 150 and 1.75 indicates a deviation in the same direction. It is concluded that Equation (1) is satisfactory for correlation but that further investigation of the value of the constants and their dependence on packing parameters is warranted.

Flow Pattern

Three modes of flow were observed with two phases. In droplet flow small droplets of the dispersed phase were

TABLE 3. COEFFICIENTS FOR ERGUN EQUATION

Packing	k_1	k_2
0.501-in. spheres	315	1.16
0.340-in. spheres	254	1.52
	349*	1.19*
0.164-in. spheres	210	1.28
Ergun (11)	150	1.75

* Obtained after column was repacked for water-isooctane tests.

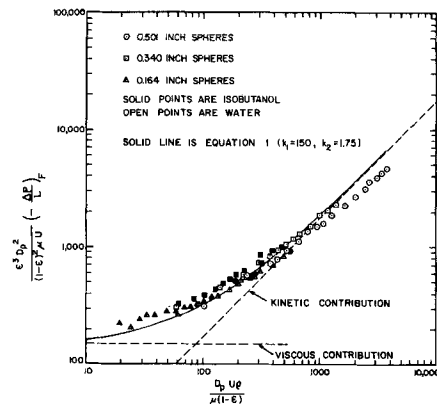


Fig. 3. Comparison of single-phase pressure gradients with the Ergun equation isobutanol-water system.

observable as in Figure 4a. In pseudohomogeneous flow no distinct droplets were observable, but the liquid mixture had a milky appearance. This regime occurred at high total flow rates, small packing diameter, and low interfacial tension. However, the enlarged photographs showed very small droplets to be present. Furthermore, very small droplets were observable when the flow was stopped. In slug flow, as also described by Larkins and White (12), a slug of liquid in pseudohomogeneous flow appeared at the entrance to the test section as indicated in Figure 4b, elongated and became irregular as indicated in Figure 4c, and sometimes disappeared before reaching the top of the bed. Separation between slugs averaged 1 ft. The slugs first appeared as a ring around the circumference and then behaved differently at different points on the circumference suggesting a continuous layer across the column.

Slug flow generally occurred at high flow rates and organic-to-water-phase flow ratios of about 3 to 1. Since the water phase preferentially wet the glass, it would be expected to be continuous. This was indeed observed at low organic phase fractions. When pseudohomogeneous and slug flow developed, it was difficult to determine which phase was continuous. Observation during the separation process immediately following actuation of the quick-closing valves did give an indication of which phase was continuous. Droplets of dispersed phases were observed to fall through the continuous phase, indicating that the heavier water phase was discontinuous for organic-to-water-phase flow ratios greater than 3 to 1. This observation is consistent with the maximum volume fraction for a dispersed phase reported by Selker and Sleicher (13) for stirred vessels.

A possible explanation for the phase reversal is that the most compact arrangement of uniform spheres, hexagonal close packing, has a void fraction of 25.95%. (A distribution of drop sizes permits a slightly greater dispersed

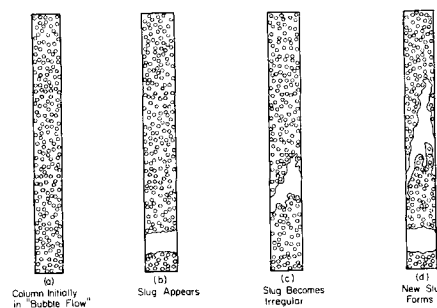


Fig. 4. Slug flow pattern.

fraction.) As the percentage of organic phase is increased, the spherical droplets of organic phase become closer and closer. At the point of touching the organic phase becomes continuous and the water phase discontinuous.

Phase Holdup

The term *phase holdup* ordinarily refers to the discontinuous phase. Because of uncertainty for some conditions as to which phase was continuous, phase holdup H is defined as the fraction of the void volume in the packed bed occupied by the nonwetting phase, in all cases herein the organic phase.

The time required to establish or re-establish the steady state holdup is important in commercial extraction as well as of intrinsic interest. The transient data which were obtained for the isobutanol water system did not follow a slug displacement curve but rather could be represented reasonably well by the exponential relationship

$$\frac{H - H_\infty}{H_i - H_\infty} = e^{-\beta t} \quad (2)$$

as illustrated in Figure 5. Insufficient data were obtained to define the dependence of the time constant on the flow rates, packing, and fluid properties.

The steady state holdup data are illustrated in Figure 6. If there were no slip, the steady state holdup would be represented by the expression

$$H = \frac{U_o}{U_w + U_o} \quad (3)$$

However the less dense (organic) liquid rises relative to the denser liquid (water). This relative motion, called the *slip velocity*, can be computed from the holdup and rate data with the expression

$$V_s = \frac{U_o}{\epsilon H} - \frac{U_w}{\epsilon(1-H)} \quad (4)$$

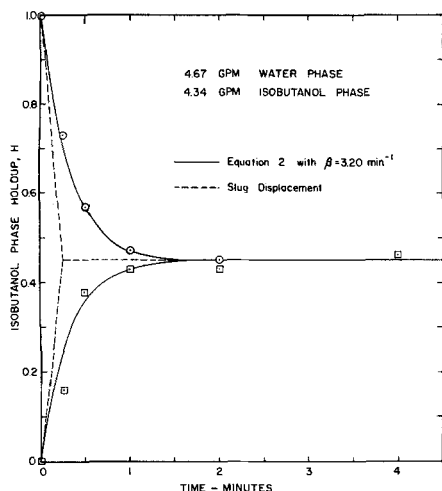


Fig. 5. Displacement of initial phase holdup, isobutanol-water system with 0.501-in. spheres.

TABLE 4. EXPONENT FOR PHASE HOLDUP

System	α
Isobutanol-water, 0.501-in. spheres	1.08
Isobutanol-water, 0.340-in. spheres	1.10
Isobutanol-water, 0.164-in. spheres	1.27
Isooctane-water, 0.340-in. spheres	1.20
Isooctane and Alkaterge C-water, 0.340-in. spheres	1.26

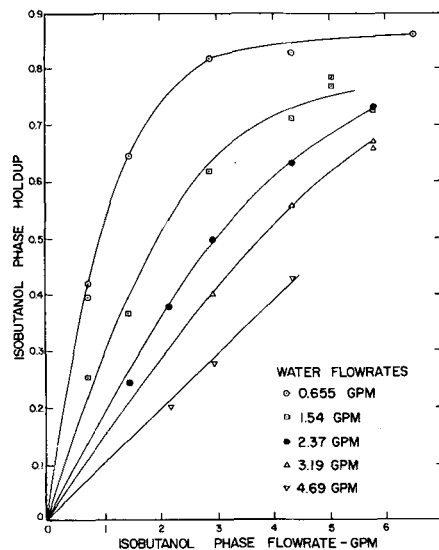


Fig. 6. Phase holdup data, isobutanol-water system with 0.164-in. spheres.

For countercurrent flow the slip velocity has been correlated successfully and used to predict holdup. However with the much larger values of U_o and U_w which were investigated in concurrent flow the slip velocity was generally a small difference between two large numbers and hence quite uncertain.

Wicks and Beckmann (14) and Markas and Beckmann (15) correlated data for countercurrent liquid-liquid systems by an equation of the form

$$H = A U_D^r + B U_D U_C^s \quad (5)$$

This expression is obviously unsatisfactory for concurrent flow, since it implies that H is unbounded for increasing flow rates. The following alternative dependence on the

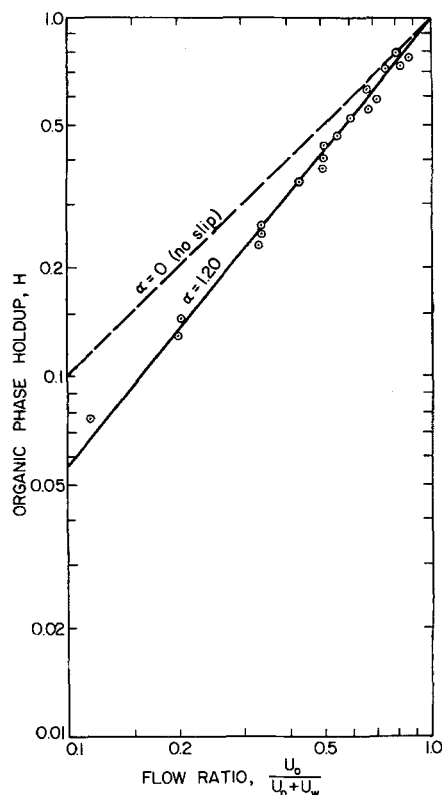


Fig. 7. Correlation of organic phase holdup data, isooctane and Alkaterge C-water system with 0.340-in. spheres.

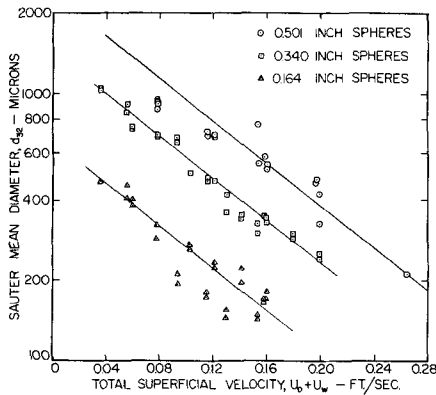


Fig. 8. Effect of velocity on Sauter mean diameter, isobutanol-water system.

power of the velocities proved to correlate the data satisfactorily:

$$H = \left(\frac{U_o}{U_o + U_w} \right)^\alpha \quad (6)$$

The exponent α was evaluated by least squares for each set of data after the logarithm of Equation (6) was taken. The correlation is illustrated in Figure 7, and the results are summarized in Table 4. The error resulting from the assumption of no slip may be quite significant at low holdup. The coefficient α (and hence the slip velocity) appears to increase with increasing density difference and increasing packing surface, as would be expected.

Drop-Size Distribution and Mean Diameter

The droplets were found to be quite spherical in shape and to have a Gaussian distribution; that is the accumulative percentage of droplets below a given size fell on a straight line on a probability scale.

The Sauter mean diameter

$$d_{32} = \frac{\sum_{j=1}^n d_j^3}{\sum_{j=1}^n d_j^2} \quad (7)$$

that is the diameter of a sphere which has the same ratio of surface to volume as the total population of spheres, is a significant criterion for mass transfer since it permits calculation of the interfacial area from the holdup. Values of the Sauter mean diameter determined from repeated runs and photographs showed a maximum deviation of about 5% from the mean. The effects of total flow rate and packing size on the Sauter mean diameter are illustrated in Figures 8 and 9. Based on the indicated effects and dimensional considerations the following general correlation was developed by least squares for all of the data:

$$\frac{d_{32}}{D_p} = 0.168 e^{-20.5 \mu_M (U_o + U_w) / \sigma} \quad (8)$$

The definition of the mean viscosity μ_M is discussed in the following section. The correlation coefficients for the equation is 0.961. The representation of the data by Equation (8) is illustrated in Figure 10.

The photographs were taken just inside the column wall. The mean drop size may be different in the interior of the packed bed owing to the change in porosity and velocity. However in view of the high degree of dispersion and the uniform appearance of the dispersion across the column at the exit from the packing, it is presumed that the same general distribution of drop size and the same general dependence on the variables occurred across the column.

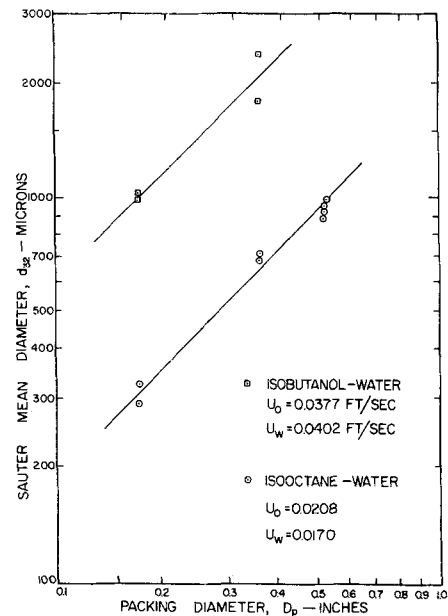


Fig. 9. Effect of packing diameter on Sauter mean diameter, isooctane-water system.

Two-Phase Gradient

Several expressions have been proposed for the pressure gradient due to gravity in two-phase systems. Larkins and White (12) used the in situ volumetric average density to calculate the pressure gradient due to gravity:

$$\rho_M = \rho_w^{(1-H)} + \rho_o^H \quad (9)$$

Hughmark and Pressburg (16) applied the Bernoulli equation separately to each phase to derive an alternative expression equivalent to the following:

$$\rho_M = \frac{\rho_w U_w + \rho_o U_o}{U_w + U_o} = \rho_w \left(1 - \frac{U_o}{U_o + U_w} \right) + \rho_o \left(\frac{U_o}{U_o + U_w} \right) \quad (10)$$

For relatively small slip velocities, such as were experienced in this work, the mean densities calculated from Equations (9) and (10) differ only slightly. Equation (10) and the following equivalent expression for the mean viscosity were used herein for correlation:

$$\mu_M = \mu_w \left(1 - \frac{U_o}{U_o + U_w} \right) + \mu_o \left(\frac{U_o}{U_o + U_w} \right) \quad (11)$$

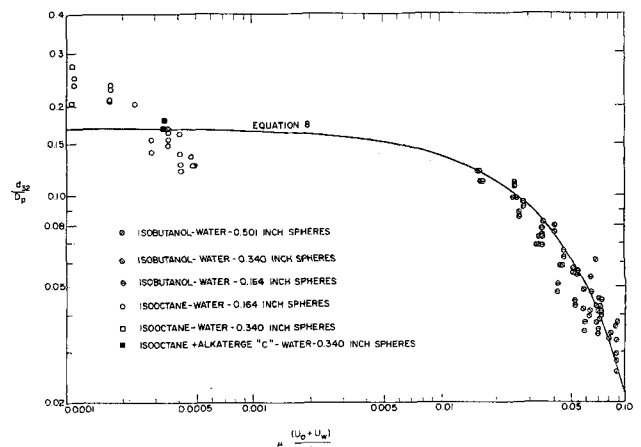


Fig. 10. General correlation for Sauter mean diameter.

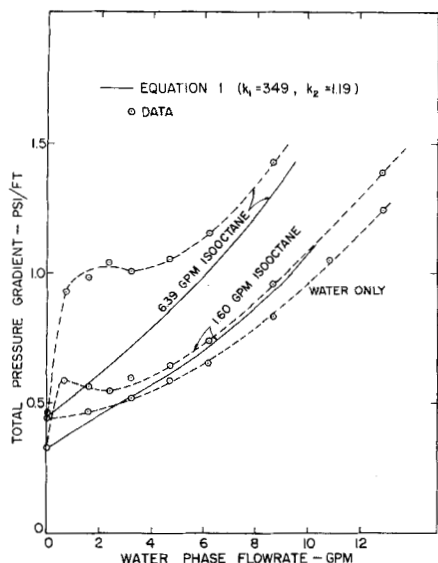


Fig. 11. Total pressure gradient data, isooctane-water system and 0.340-in. spheres.

In Figure 11 the measured pressure gradient is compared with that predicted by Equation (1) [from the values of k_1 and k_2 given in Table 3 for this packing] plus the static gradient corresponding to Equation (1). The measured values necessarily approach the predicted values at both extremes of phase ratio but demonstrate large deviations at intermediate phase ratios. The cross over in the experimental curves for 0 and 1.60 gal./min. of isooctane occurs because the static gradient for pure water exceeds the static plus dynamic gradient for 1.60 gal./min. of pure isooctane.

The ratio of the pressure gradient due to friction to the pseudo-single-phase value given by Equation (1) can be expressed as

$$\Phi = \frac{(-\Delta P/L)_F}{(-\Delta P/L)_{F, \text{Equation (1)}}} = \frac{(-\Delta P/L)_T - \rho_M}{(-\Delta P/L)_{F, \text{Equation (1)}}} \quad (12)$$

with ρ_M and μ_M calculated from Equations (10) and (11) and the appropriate value of k_1 and k_2 taken from Table 3. The deviation of Φ from unity must be attributable to interaction between the phases. This ratio is plotted vs. the phase ratio for the water-isobutanol system in Figure 12. The data scatter considerably since each point represents a different total flow rate, but a maximum is quite apparent at an organic phase holdup of approximately

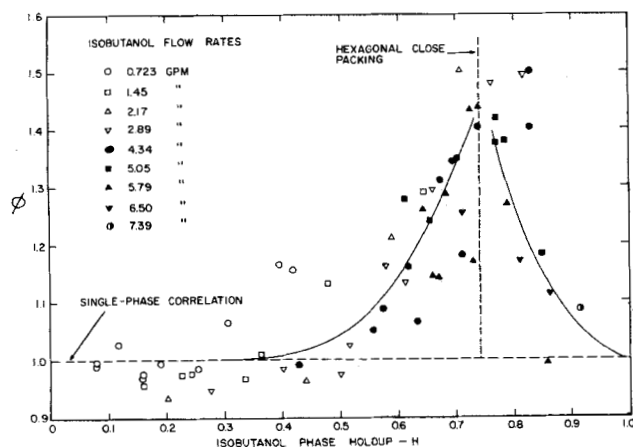


Fig. 12. Dependence of pressure gradient on phase holdup, isobutanol-water system with 0.164-in. spheres.

0.75. It was previously noted that phase reversal occurred at approximately this composition.

Dimensional considerations and the observed dependence on total flow rate, surface tension, and packing diameter then led to the development of the following empirical equation for the water-isobutanol and water-isooctane systems:

$$\Phi = 0.723 \left[\frac{\sigma}{\rho_M D_p (U_O + U_W)^2} \right]^{0.624} e^{-5.59(H-0.75)^2} \quad (13)$$

The constant 0.75 was chosen on the basis of Figure 12 and the previously discussed considerations. The other three constants were evaluated by least squares. The use of four constants was required by the shape of the curve in Figure 12 rather than by the precision of the data, and no physical significance should be attributed to the values of the three arbitrary constants. The data upon which the correlation is based (open symbols) are compared with Equation (13) in Figure 13. Because of congestion only every tenth point is plotted for predicted values of Φ less than 1.1. 90% of the data are within the $\pm 20\%$ limits.

The measured pressure gradients for the water-isooctane system with the addition of Alkaterge C were essentially the same as without. Hence if the data for Alkaterge C were plotted in Figure 13 in terms of the measured interfacial tension, the points would fall far to the left of the 45-deg. line. However, when these data are plotted in terms of the unaltered interfacial tension for water and pure isooctane, the agreement is somewhat better although the scatter is greater than for the other systems as indicated in Figure 13. It is concluded that the surfactant produces much less change in the effective interfacial tension of dynamic, dispersed systems than indicated by the static measurement. During the measurement with the ring tensiometer the surfactant concentrates at the interface; in the highly dispersed state the concentration at the interfaces is less because of the increased and variable ratio of surface to volume.

Re-examination of the correlation for the Sauter mean diameter (Figures 9 and 10) indicates that use of the unaltered interfacial tension would not shift the points for water and isooctane plus Alkaterge C significantly in terms of the scatter of the data. The variation of the exponent α in Equation (6) is also within the uncertainty of the data.

Larkins and White (12) attributed the increased pressure gradient for the concurrent flow of gas and liquid through a packed bed to the irreversible work of compression performed on the gas phase. This explanation is obviously not applicable to liquid-liquid flow. Cengel et al.

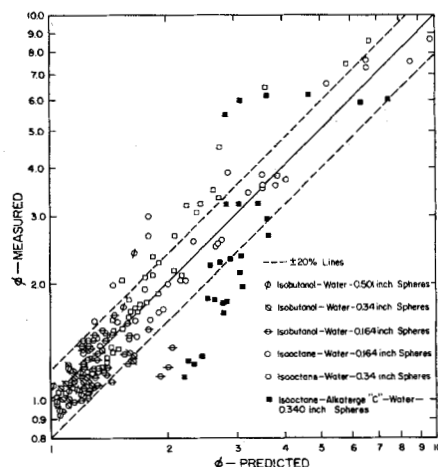


Fig. 13. Comparison of measured and predicted pressure gradients.

(17) attributed the increased pressure gradient for the flow of liquid dispersions through open pipe to an increase in the viscosity. However the effective viscosity proposed by Baron, Sterling, and Schueler (18) did not prove effective in correlating the data obtained herein.

Surface energy effects may be the major contributor to the increased pressure gradient. The energy required to form a dispersion appears as a pressure loss. When coalescence occurs, some of the surface energy is presumably converted to thermal energy rather than recovered as pressure. The stretching of droplets to pass through small openings in the packing, the Jamin effect (9), results in the further conversion of pressure energy to surface energy to thermal energy. The pressure loss associated with the formation of the observed dispersions is calculated to be approximately 12% of the measured pressure loss per foot of packed bed. This calculation is very conservative since repeated stretching, coalescence, and dispersion undoubtedly occur within that distance. The maximum in the pressure gradient appears to accompany the phase reversal. As the organic phase becomes continuous, the interfacial area would be expected to decrease and the pressure loss due to surface formation would be expected to decrease.

CONCLUSIONS

The conclusions drawn from the data and correlations may be summarized as follows.

1. The form of the Ergun equation is satisfactory for the correlation of the data for single-phase flow through packed beds of spheres, but the two coefficients vary with the packing arrangement.

2. A mixture of immiscible fluids flowing through a packed bed cannot be treated as a pseudo single phase with effective properties. The large increase in pressure gradient appears to be associated with surface formation. Phase inversion and a maximum in the pressure gradient occur at an organic phase holdup of approximately 75%. The observed pressure gradients can be represented reasonably well for all conditions by Equation (13).

3. The slip velocity is small but measurable, and the phase holdup can be represented by Equation (6).

4. The initial-phase holdup appears to decay exponentially rather than linearly with displacement.

5. The dispersed phase consists of spherical droplets with a Gaussian distribution of diameters. The Sauter mean droplet diameter is directly proportional to the diameter of the packing and can be correlated with velocity, viscosity, and interfacial tension as indicated by Figure 10.

6. The addition of a surfactant which reduced the interfacial tension by a factor of 3 did not affect the pressure gradient or droplet formation significantly.

ACKNOWLEDGMENT

The authors gratefully acknowledge the contribution of iso-octane by the Phillips Petroleum Company and isobutanol by the Union Carbide Chemicals Company. The suggestions of Professors E. F. Brater, M. R. Tek, G. B. Williams, and J. L. York were invaluable. Fellowship support was provided Robert G. Rigg by the National Science Foundation. Much of the equipment was adapted from that constructed by R. P. Larkins.

NOTATION

A, B = arbitrary constants, Equation (5)
 D_p = diameter of packing, length
 d = diameter of a droplet, length
 g = acceleration due to gravity, length/time²
 H = fractional holdup of nonwetting phase in void volume

k_1, k_2 = arbitrary constants, Equation (1)
 L = packed length, length
 n = number of droplets in photograph
 p = pressure, mass/(length)(time)²
 r, s = arbitrary constants, Equation (5)
 t = time after introduction of second liquid phase, time
 U = superficial velocity, length/time
 V_s = slip velocity [see Equation (4)], length/time

Greek Letters

α = arbitrary constant, Equation (6)
 β = arbitrary constant, Equation (2), time⁻¹
 ϵ = void fraction of packed bed
 μ = viscosity, mass/(length)(time)
 ϕ = dimensionless pressure gradient [see Equation (12)]
 ρ = density, mass/length³
 σ = interfacial tension of liquid phases, mass/time²

Subscripts

D = dispersed phase
 C = continuous phase
 O = organic phase
 W = aqueous phase
 F = due to friction
 T = total
 i = initial
 ∞ = steady state
 M = mean
 32 = Sauter mean [see Equation (7)]

LITERATURE CITED

- Leacock, J. A., and S. W. Churchill, *A.I.Ch.E. Journal*, **7**, 196 (1961).
- Rigg, R. G., Ph.D. thesis, The University of Michigan, Ann Arbor, Michigan (1963).
- Seidell, A., "Solubilities of Organic Compounds," 3 ed., Vol. 2, p. 268, Van Nostrand, New York (1941).
- Phillips Petroleum Co., private communication.
- Hodgman, C. D., ed., "Handbook of Chemistry and Physics," 36 ed., Chemical Rubber Publishing Co., Cleveland, Ohio (1954).
- Perry, J. H., ed., "Chemical Engineers Handbook," 3 ed., p. 192-3, McGraw-Hill, New York (1950).
- Lange, N. A., ed., "Handbook of Chemistry," 9 ed., Handbook Publishers Inc., Sandusky, Ohio (1956).
- "International Critical Tables," Vol. 4, p. 436, McGraw-Hill, New York (1926).
- Hassan, M. E., R. F. Nielsen, and J. C. Calhoun, *J. Petrol. Technol.*, **5**, 299 (1953).
- Ergun, S., and A. A. Orning, *Ind. Eng. Chem.*, **41**, 1179 (1949).
- , *Chem. Eng. Progr.*, **48**, 89 (1952).
- Larkins, R. P., and R. R. White, *A.I.Ch.E. Journal*, **7**, 231 (1961).
- Selker, A. H., and C. A. Sleicher, Jr., Paper presented at 50th National Meeting of A.I.Ch.E., Buffalo, New York (1963).
- Wicks, C. E., and R. B. Beckmann, *A.I.Ch.E. Journal*, **1**, 426 (1955).
- Markas, S. E., and R. B. Beckmann, *ibid.*, **3**, 223 (1957).
- Hughmark, G. A., and B. S. Pressburg, *ibid.*, **7**, 677 (1961).
- Cengel, J. A., A. A. Faruqui, J. W. Finnigan, C. H. Wright, and J. G. Knudsen, *ibid.*, **8**, 335 (1962).
- Baron, T., C. S. Sterling, and A. P. Schueler, "Proceedings Third Midwestern Conference on Fluid Mechanics," p. 103, University of Minnesota Press, Minneapolis, Minnesota (1953).
- Scheidegger, A. E., "The Physics of Flow Through Porous Media," University of Toronto Press, Toronto, Ontario (1960).

Manuscript received September 19, 1964; revision received June 3, 1964; paper accepted July 10, 1964.