The Mechanics of Vertical Moving Liquid-Liquid Fluidized Systems: I. Interphase Contacting of Droplets Passing Through a Second Quiescent Fluid

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Previous publications have shown that for solid spheres fluidized in water a unique relationship exists between the slip velocity and the system holdup. With this work as a model a method is now presented for estimating the behavior of the liquid-in-liquid spray column in which droplets of one phase move through a second quiescent phase.

Combining the solids fluidization results with information on the single droplet terminal velocity one can obtain a design estimate of the holdup or interphase contact area for the liquid-liquid spray column. This design estimate includes the particular nature of the liquid droplet of being susceptible to internal circulation, oscillation, and distortion.

The effective contact area between two phases directly affects the over-all rate of any process occurring across the phase boundary. In most transfer operations, where one fluid is dispersed through the other, this area can at present be estimated or the system holdup determined only by a difficult direct measurement. Consequently most earlier studies of transfer processes have combined the contact area with the intrinsic rate characteristics of the system.

In a previous publication from this laboratory (*12*) a detailed theoretical analysis was presented for predicting the behavior of all types of vertical moving fluidized systems. The basic postulate of this development was the proposal that a single unique relationship exists between the slip velocity and the holdup for a system comprising one particle size. For such a system the holdup is related directly to the interfacial area. Subsequent publications (*14, 15, 17*) have tested this theory with rigid glass spheres fluidized by water. Under all vertical-flow arrangements the agreement between theory and experiment was excellent thereby substantiating the single unique relationship mentioned above.

The fluidized systems thus studied may be characterized as ideal in the sense that they involve uniform dispersed particles which are rigid and impermeable. Particulate fluidization always occurs under these operating conditions. Of paramount importance is the extension of the concepts developed for ideal systems to the area of nonideal systems. This paper reports on the behavior of the liquid-in-liquid spray column in which droplets of one phase move through a second quiescent continuous phase. In contrast to the previous ideal systems with solid spheres, the liquid droplets are subject to internal circulation, oscillation, and distortion as they pass through the second fluid.

The present experimental results demonstrate that the holdup may be related directly to a slip-velocity ratio, for example a ratio of the normal slip velocity between the two phases in the column to the terminal velocity of a representative single droplet. This relationship is a generalization of the previously defined unique slip velocity-holdup function for solid particles.

Holdup data are presented for the systems methyl isobutyl ketone-water, isobutanol-water, toluene-water, and isoamyl alcohol-water operating in a 1-⅞-in. Elgin type of spray tower. In each case the organic solvent is the dispersed phase. The holdup was measured directly in the tower as a function of the system parameters by sealing off a part of the tower proper. The system parameters included the dispersed phase flow rate, the droplet size in the column, and the physical properties of the organic solvents. Detailed data are also presented to show that the droplet size distribution in the tower is normal in the Gaussian sense.

BACKGROUND INFORMATION

Since the present liquid-in-liquid system compares with the familiar batch fluidization of solids, an extensive survey was made of the present status of this latter type of operation. In addition to the normal experimental and empirical approach a number of models have been proposed in which the fluidized state is treated as an ideal gas (*20*), as a liquid (*3*), or in terms of statistical interactions (*16*). To the present day however the voluminous experimental information has defied a simple analytical description.

Direct resort to the data itself, codified in various dimensionless number presentations, appears the most reliable method of obtaining an indication of the holdup of solid particles under different flow conditions. The compilation of Zenz (*21*), illustrated in Figure 1, is based on a good cross section of the published studies; this convenient description in three parameters, representing the particle size, velocity, and holdup, has been the source of the fluidization information used in the present study.

The fact that liquid droplets passing through a second continuous fluid are subject to various degrees of nonideality, that is induced internal circulation, distortion, and oscillation of the drop-

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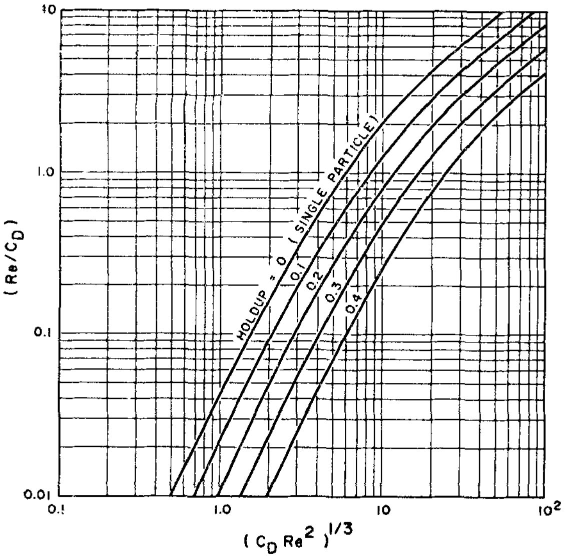

Fig. 1. Zenz correlation for fluidized solids.

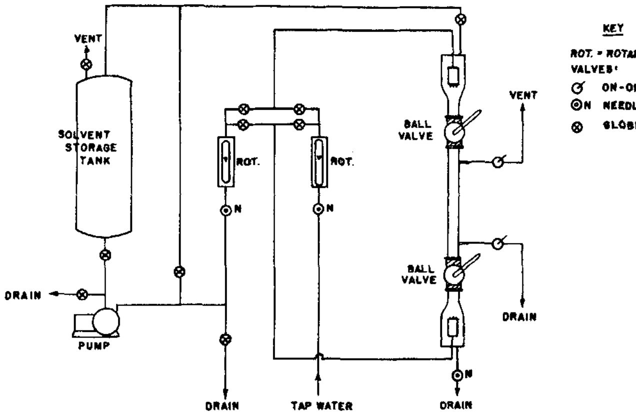

Fig. 2. Schematic diagram of experimental apparatus.

lets, has been described by a number of authors (*1*, *4*, *5*, *7*, *8*, *11*, *13*, *18*). Specific consideration of these phenomena by Calderbank and Korchinski (*2*) has given an indication that the pertinent dimensionless criteria are the Reynolds and Weber numbers along with the conventional drag coefficient. Klee and Treybal (*10*) confirmed this in their investigation; they were able to obtain a general correlation in these terms. The fall velocity of the liquid droplets reported by these authors as a function of the drop diameter shows two clear regimes. These can be described analytically in terms of the above mentioned groups, but the hump or maximum found to exist in the transition zone between the two regimes has not been amenable to an analytical description. When presented in terms of the drag coefficient vs. the Reynolds number the Klee-Treybal data result in a family of curves which are functions of the surface tension. These curves indicate that the effects of internal circulation, droplet oscillation, and distortion (significant particularly for the larger drop sizes and high Reynolds numbers) are more pronounced the lower is the interfacial tension of the system. As a result the experimental drag coefficient curves cross each other.

EXPERIMENTAL

Apparatus

The equipment for the present study was designed to permit precise measurements of the holdup in the operating liquid-liquid spray tower. Other variables defining the process and requiring measurement included the flow rates of both phases, fluid system properties, operating temperature, and the droplet size in the tower. The particular liquid-liquid systems consisted in each case of water with one of several organic solvents; these included methyl isobutyl ketone, isobutanol, toluene, and isoamyl alcohol. The solvents were chosen to provide a representative range of the pertinent physical properties: density, viscosity, and interfacial tension.

Figure 2 shows a schematic view of the over-all apparatus and Figure 3 a diagram of the tower proper. The end sections of the glass extraction tower were constructed in the accepted Elgin design with 16-deg. conical frustra leading to the expanded ends. These sections were connected to

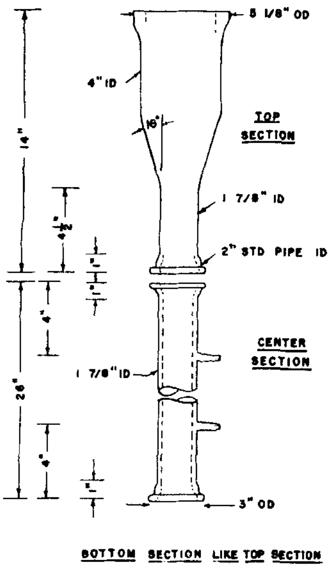

Fig. 3. Schematic diagram of glass column.

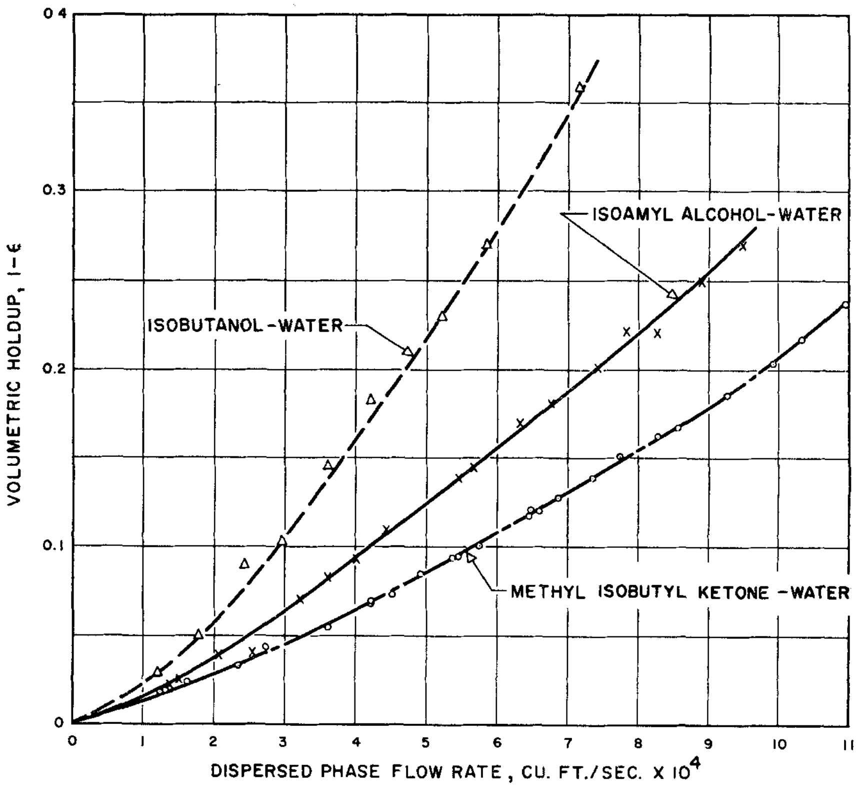

Fig. 4. Experimental holdups vs. dispersed-phase flow rate, 1/8-in. nozzles.

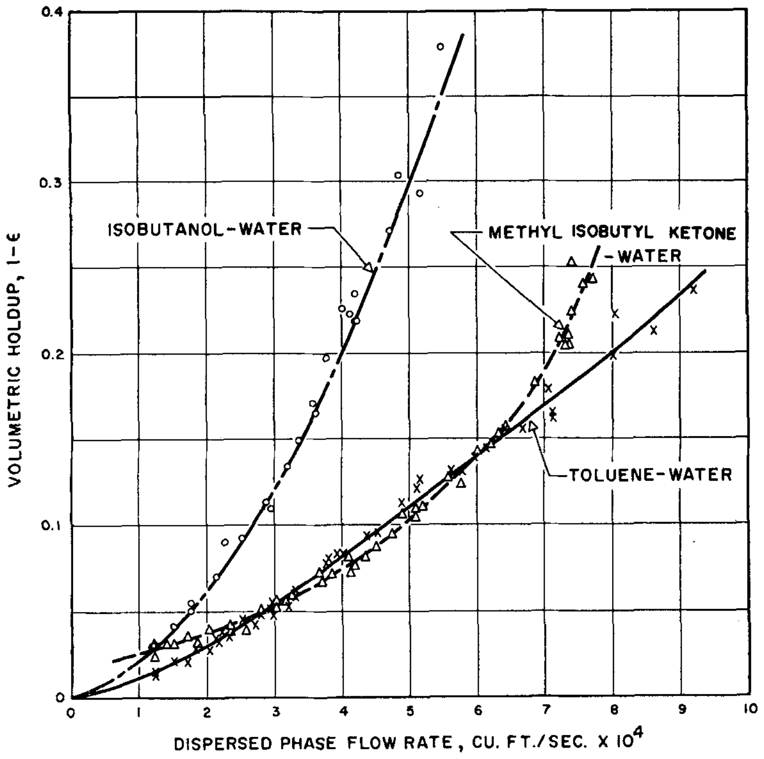

Fig. 5. Experimental holdups vs. dispersed-phase flow rate, 1/16-in. nozzles.

the center section through two quick opening ball valves. When open the valves offered an unobstructed uniform cross-section path through the entire tower. Holdup in the central section could be directly measured upon simultaneously closing these two valves and allowing the two phases to separate. Two condenser taps along the length of the center section were used in venting and provided sample taps during column operation.

Both phases were supplied to the column through the standard type of nozzle heads. In all cases the organic solvent was the dispersed phase. Each head was packed with ¼-in. glass rings to circumvent channeling and allow a uniform delivery through all the nozzles. The nozzles themselves, ⅛ and 1/16 in. diameter, were of a sharp edged variety and were machined from a single diameter brass rod so as to be interchangeable in number and size.

Drop sizes and shapes were recorded photographically for representative runs during the operation of the tower. Correction was made for the distorting effect of refraction through the curved column wall. The correction was significant for droplets larger than 3 mm. diameter. The physical properties and the operating-tower temperature were measured in the samples tapped from the tower during the course of the experimental run. The properties used to describe the particular systems were density, viscosity, and surface tension. These were measured by the pycnometer, the Ostwald viscosimeter, and Harkins drop-weight method (*6*), respectively. These measurements were reproducible with no change evident during the course of the experiments. In all cases the liquids used were mutually saturated. Table 1 summarizes the experimentally determined results.

PROCEDURE AND OBSERVATIONS

The experimental procedure used to establish the relationship between the slip velocity and the holdup in the liquid-liquid spray tower included the following:

TABLE 1. TABULATION OF PHYSICAL PROPERTIES

Key:
Visc. = viscosity at 25°C. in centipoises
Dens. = density at 25°C. in g./cc.
I. T. = interfacial tension with water at 25°C., dynes/cm.

Liquid	Visc.	Dens.	I. T.
Methyl isobutyl ketone	0.8252	0.7995	8.82
Isobutanol	4.2789	0.8185	2.08
Toluene	0.7158	0.8595	30.3
Isoamyl alcohol	4.8812	0.8220	4.58

1. After steady state was reached in each run, the flow rate of the organic phase was measured by a rotameter. Throughout the present work a static continuous phase was used.
2. For a representative number of runs photographs were taken to be used subsequently in determining the droplet sizes.
3. Samples were tapped to obtain an indication of the temperature in the operating tower and to be used for physical property measurements.
4. The ball valves were then quickly closed, isolating the central section of the tower; after settling the volume of the dispersed phase (that is the holdup) was measured.

The following general observations were made during the experiments:

1. The data were all taken between 22° and 26°C., and no meaningful temperature effect could be detected in this narrow range.
2. The character of the droplet distribution did not change noticeably along the

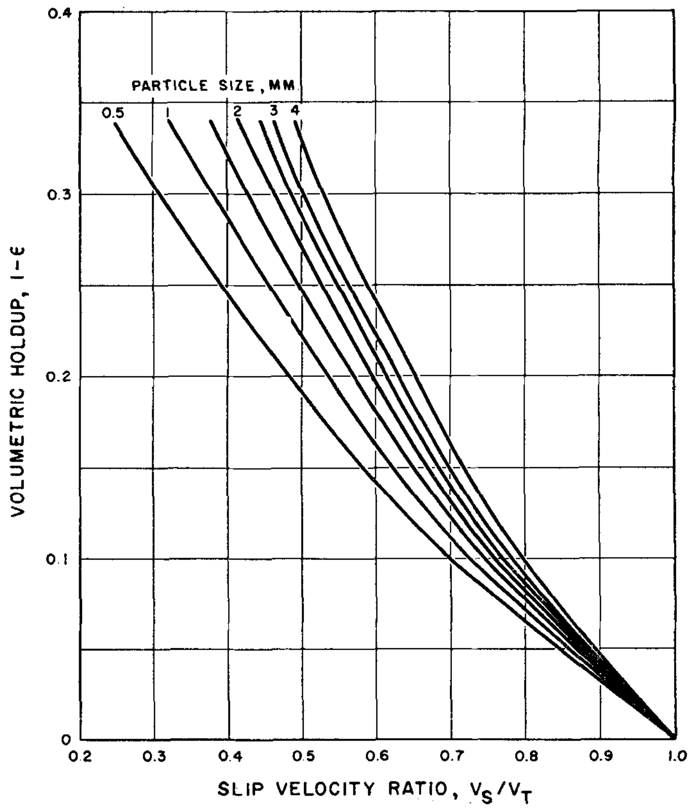

Fig. 6. Batch expansion curve for a system of rigid spheres of density equal to isobutanol as predicted by the Zenz correlation.

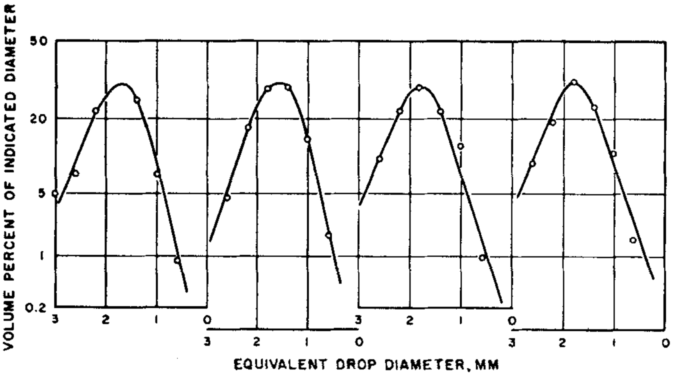

Fig. 7. Drop-size distributions for isobutanol in water, 1/8-in. nozzles.

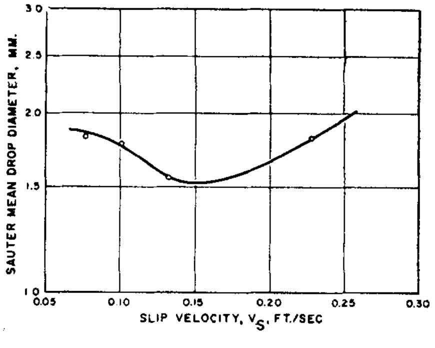

Fig. 9. The Sauter mean diameter vs. the slip velocity. Isobutanol in water, 1/8-in. nozzles.

length of the center section of the tower. No droplet coalescense in the tower proper was observed, although some foaming was observed at the terminals in several of the high-holdup runs of the methyl isobutyl ketone-water systems.

3. The droplets were somewhat distorted from the spherical into oblate ellipsoids while moving through the tower. However, as earlier workers have successfully treated their data in terms of the Sauter mean diameter, the measured droplet dimensions here have also been converted into this form. The Sauter mean is simply the diameter of a sphere of the same volume as the distorted droplet.

EXPERIMENTAL RESULTS

Figures 4 and 5 present (in the form of disperse-phase holdup vs. the disperse-phase flow rate) the experimental data collected in the present investigation. Figure 4 shows data using the ⅛-in. nozzles and Figure 5 the 1/16-in. nozzles. The reproducibility of these data is excellent. In the discussion which follows the data for the isobutanol-water system with ⅛-in. nozzles will be specifically considered.

TABLE 2. DROP-SIZE MEASUREMENTS AND DISTORTION FOR SYSTEM OF ISOBUTANOL-WATER—⅛-IN. NOZZLES

Volume percentages in the size range indicated by its midpoint

Run no.	1	2	3	4
Mean size of droplets (mm.)				
0.6	1.0	1.8	1.6	0.9
1.0	13.1	14.9	11.7	7.9
1.4	22.5	30.8	23.2	26.0
1.8	30.2	30.3	35.2	30.1
2.2	23.1	17.5	18.7	22.3
2.6	10.1	4.7	9.6	7.9
3.0				5.0
Distortion				
$(a/b)_m$	0.805	0.849	0.824	0.856

Mean size of droplets given in terms of Sauter-mean diameters of individual droplets.

a and b are characterizing dimensions of droplets. The distortion is the volume-weighted average of the individual droplets a/b values.

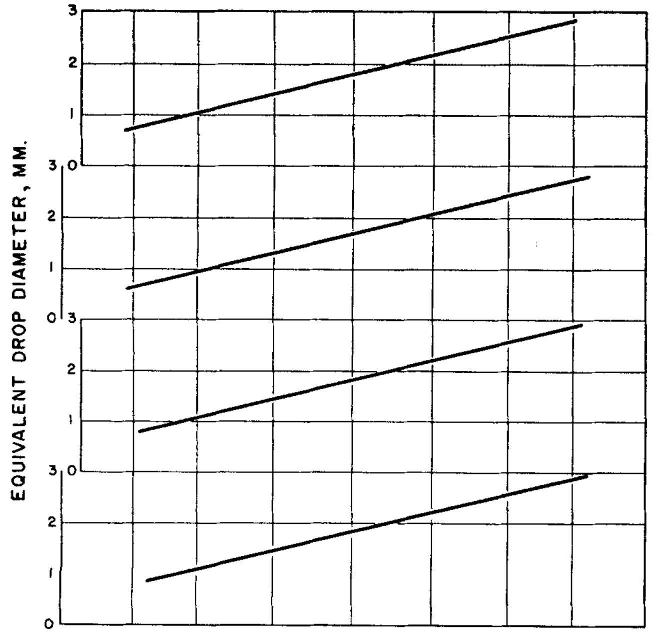

Fig. 8. Cumulative drop-size distribution for isobutanol in water, 1/8-in. nozzles.

DISCUSSION

The model proposed in this paper assumes that, for both liquid and solids dispersed systems, a given holdup will define a unique slip-velocity ratio, V_S/V_T. This ratio measures the approach of the slip velocity in the tower to the terminal velocity for the single droplet or particle. The unique ratio proposed is an extension of the previous relationship for ideal systems but one which now takes into account the nonideal flow behavior of liquid-liquid fluidized systems. In order to confirm or to disprove this model the present liquid-liquid data will be compared with the data for an analogous system of batch fluidized solid particles.

BEHAVIOR OF SOLID PARTICLES ANALOGOUS TO LIQUID DROPS

The Zenz correlation may be used to provide an estimate of the holdup-slip velocity ratio relationship for a solid particles fluidized system. When one assumes that solid spheres of density equal to that of isobutanol are fluidized by water, this correlation yields the data represented graphically in Figure 6.

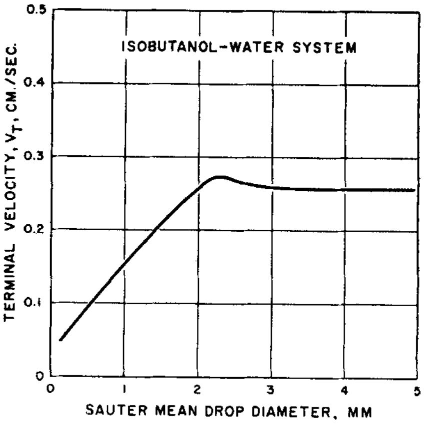

Fig. 10. Terminal velocity of isobutanol droplets in water as predicted by the Klee and Treybal correlation.

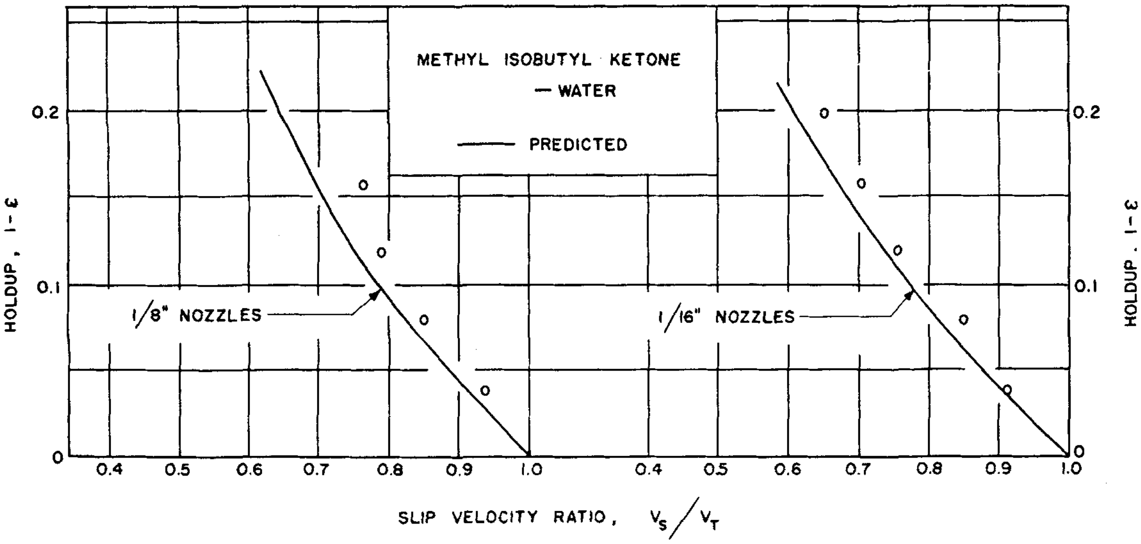

Fig. 11. Observed vs. predicted slip velocity-holdup curves for methyl isobutyl ketone water, 1/16-and 1/8-in. nozzles.

In this chart the holdup is plotted vs. the slip-velocity ratio for a range of spherical particle diameters. These curves correspond to the behavior of hypothetical rigid isobutanol spheres fluidized by water.

DROPLET SIZE DISTRIBUTION IN TOWER

Visual observations of the droplets passing through the tower suggests that a distribution of droplet sizes exists at any given tower cross section. Since the theoretical fluidization analysis previously referred to considers in each case all particles present in the system to be of the same fixed particle size, it is necessary to obtain a mean diameter for the spectrum of droplet sizes which occurs with the liquid in liquid systems studied here. A large number of photographs were taken of the tower as the disperse droplets passed through the second continuous fluid. Detailed measurements of the droplets in these photographs were then used to obtain droplet size distributions. Figure 7 shows the result of such measurements for the isobutanol-water system with 1/8-in. nozzles and is typical of all of the systems studied. The data are shown for four representative experimental runs with different flow rates used and the Sauter mean diameter of the droplets plotted vs. the volume fraction corresponding to the diameter indicated. The Sauter mean diameter is the diameter of a sphere of the same volume as the distorted droplet. Table 2 presents the data used in Figure 7 plus the actual distortion measurements of the droplets. From these curves the Sauter drop diameters were then plotted vs. the cumulative distributions in Figure 8 with probability paper used. It is notable that straight lines which are characteristic of a normal or Gaussian distribution describe the data. Furthermore uniform slopes are obtained indicating a constant variance of the distributions. This makes it possible to describe the effect of velocity on the droplet size in a given system by means of a single size parameter and the uniform slope. When one takes the 50% mean diameter of the cumulative distribution as the appropriate size parameter, the slip velocity of the system can be calculated as a function of the drop size. The data thus calculated are shown in Figure 9. Figure 9 is typical but represents only a small portion of the data available for this system and for the other liquid-liquid

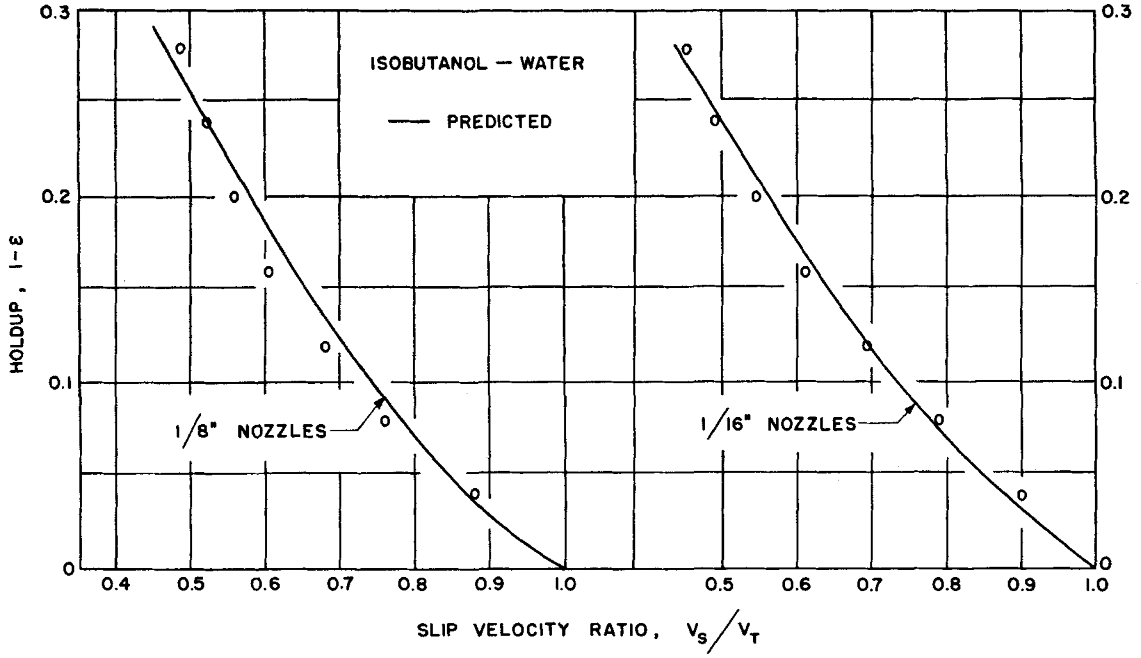

Fig. 12. Observed vs. predicted slip velocity-holdup curves for isobutanol water, 1/16-and 1/8-in. nozzles.

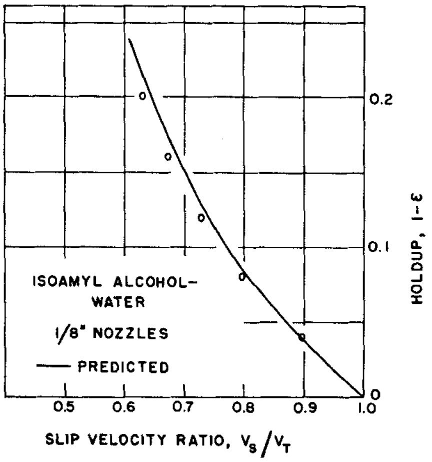

Fig. 13. Observed vs. predicted slip velocity-holdup curves for isoamyl alcohol water, 1/8-in. nozzles.

systems investigated in this study (*19*). In all cases the results are comparable.

MEAN DROPLET TERMINAL VELOCITIES

From the data of Keith and Hixson (*9*) and from the correlation of Klee and Treybal the terminal velocity of single droplets of isobutanol in water can be calculated as a function of the Sauter drop diameter. Figure 10 shows a plot of some of these data. Since the present data, with a mass of droplets passing through the column, exhibit droplet size distribution, there is no one single terminal velocity for all droplets in the system. A mean droplet terminal velocity can however be calculated with the volume weighted mean sizes used. Specifically the 10-30-50-70-90% points of the size distributions can be selected and the corresponding terminal velocities averaged to yield a mean terminal velocity for the mass of isobutanol droplets.

COMPARISON OF LIQUID-DROPLET AND SOLID-PARTICLE HOLDUPS AS A FUNCTION OF THE SLIP-VELOCITY RATIO

There are now sufficient data and information available to allow a direct comparison to be made between the actual liquid-droplet holdups and the hypothetical holdups which would exist in a solid-particle system of the same density as a function of the slip-velocity ratio. To illustrate the technique a holdup of $(1 - \epsilon) = 0.200$ has been experimentally measured for the isobutanol-water system. From Figure 4 this holdup corresponds to a flow rate of 4.66×10^{-4} cu. ft./sec. Since the tower cross section is 1.92×10^{-2} sq. ft., the slip velocity can be calculated to be

$$V_s = \frac{(4.66 \times 10^{-4})(0.200)}{1.92 \times 10^{-2}} = 0.121 \text{ ft./sec.}$$

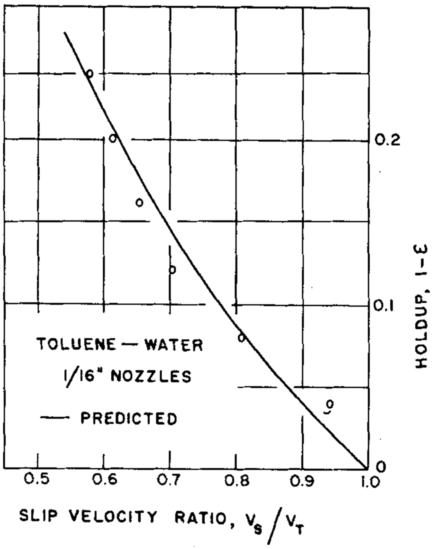

Fig. 14. Observed vs. predicted slip velocity-holdup curves for toluene water, 1/16-in. nozzles.

From Figure 9 the Sauter mean droplet diameter under these conditions is 0.162 cm. Using this as the 50% point of the drop distribution and constructing a line parallel to those in Figure 8, one can obtain an estimate of the distribution in terms of the 10-30-50-70-90% drop dimensions. The appropriate terminal velocities for each of these can be found from Figure 8. An averaging of the results yields the mean terminal velocity of 0.217 ft./sec. The slip-velocity ratio based upon experiment is then

$$V_S/V_T = 0.121/0.217 = 0.558$$

From Figure 6 it can be seen that a slip-velocity ratio of 0.558 produces a fractional holdup of 0.215 for the solid-particle fluidized system. In this example the difference between the experimentally measured holdup of 0.200 and that predicted for the system from the fluidization data for solid particles is 7.5%.

The result of similar calculations for the six experimental systems studied are shown graphically in Figures 11 to 14. The results predicted on the basis of the solid-particle model agree with the experimental liquid-liquid results with an over-all average deviation of 11%. In view of the fact that the parent information on the solids behavior and for the single liquid droplet terminal velocities are themselves subject to departures of about this order of magnitude, these results may be taken as confirmation of the basic model. Thus it is possible to predict the holdup (and from this the contact area) in a liquid-liquid spray tower on the basis of solid-particle fluidization results.

POSSIBLE DISCREPANCIES IN THE MODEL

Two possible sources of departures of the experimental data from the predicted behavior of the rigid-particle analogue are most prominent: the shortcomings of the model for treating the methyl isobutyl ketone systems and the tendency in all the systems for the liquid-droplet holdups to curve more rapidly with increasing slip-velocity ratio than do their solid particle counterparts.

Difficulty would be expected in treating the methyl isobutyl ketone-water system for two reasons. The terminal velocities reported for this system by the several investigators cited were in distinct disagreement where they overlapped. Further the nozzles used in the present study produced droplets which partly fell into the region of Treybal's "critical diameter," where the correlation could not reliably supplement the lack of experimental data. Although no change in the interfacial tension of the system could be detected, foam was also observed to develop for this particular system at the higher holdups. No coalescense occurred in the main section of the column, but the foam did appear to promote its occurrence along the walls of the entrance region. Any influence which these factors might have had could explain departures in the directions noted.

The main weakness of the present model is the apparent tendency of all the experimental data to curve somewhat more than the predicted behavior would indicate should be the case. It is difficult to judge the significance of this tendency, but a drift toward higher slip velocities at the higher holdups seems persistent. Among the phenomena which could be responsible for the presumably faster slip of the droplets here would be their involvement in gross circulation patterns (since liquid densities are actually less than those of any solid particles studied). Such circulation was observed; however without evidence as to its occurrence in the corresponding solids systems upon which the predicted behavior was based, one cannot judge the extent of the net effect. The low-density liquid droplets are also more subject to the virtual mass effect than are the systems upon which the fluidization correlations are based. The virtual mass concept one recalls is involved with accelerated (or decelerated) motion when the mass of the displaced fluid relative to that of the moving body is significant. Qualitatively virtual mass considerations would lead one to expect relatively higher slip velocities at the higher holdups in the spray column. No effect would be expected in the more uniform motion obtained at very low holdups. At the present time one is not in a position to be more specific than this.

Future publications from this laboratory will consider the effect of the continuous-phase flow on the present results as well as an extension into the area of the gas-in-liquid spray tower.

ACKNOWLEDGMENT

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NOTATION

$1 - \epsilon$ = volumetric holdup
V_S = slip velocity, ft./sec. = vectorial difference between the average discontinuous fluid velocity and the average continuous velocity.
V_T = droplet terminal velocity, ft./sec.
Re = Reynolds number
C_D = drag coefficient

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Reaction Rates in the Synthesis of Ammonia

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A study has been made of the reaction rates of mixtures of hydrogen and nitrogen to form ammonia over a doubly promoted iron catalyst at 400° and 450°C. and at pressures up to 1,000 atm. In this work particular care has been taken to obtain data representing the true kinetics of the reaction. The reactor used was essentially isothermal, and the effects of diffusion have been reduced to a minimum. The results have been correlated by the use of the mechanism proposed by Temkin and Pyschev (*19*) with moderate success.

The kinetics of the ammonia synthesis reaction have been the subject of many investigations, and results have been published by Adams and Comings (*1*), Uchida and Kuraishi (*21*), Bokhoven and van Raayen (*3*), Siderov and Livshits (*17*, *18*), Nielsen (*14*), Emmett and Kummer (*6*), Almquist and Crittenden (*2*), and Larson and Tour (*13*). Of these investigations only three (*1*, *17*, *18*) make a claim to an essentially isothermal catalyst bed; the others report temperature variations in the bed, running as high as 15° in one case (*14*). Siderov and Livshits (*18*) report the highest pressure (500 atm.), and in no case has the entire diffusion problem been analyzed, although Bokhoven and his associates (*3*, *8*) have done valuable work on the question of diffusion within the catalyst pores, and Adams and Comings (*1*) report a calculation for the effect of diffusion between the bulk of the gas and the surface of the catalyst.

APPARATUS

Figure 1 shows the general flow diagram for the process. The mixture of hydrogen and nitrogen was compressed over mercury in the *U*-tube made from two 3-liter vessels, *E*. The pump, *B*, pumped oil into one leg of the *U*-tube and could develop a pressure of 20,000 lb./sq. in. The compressed synthesis gas was stored in the two 3.5-liter vessels, *H*; these vessels could store enough gas for about a week of continuous running at about 400 atm. pressure in the reactor.

During a run the gases passed from the storage vessels, *H*, through the clean-up train (*14*, *13*, *8*, *12*, *11*) to the reactor, *R*. This train consisted of a 1-liter vessel filled with sodium hydroxide pellets and activated carbon; here carbon dioxide and possibly oil mist were removed. Next came a 300 cc. vessel containing finely divided copper maintained at 300°C.; the copper was made from the reduction of copper hydroxide in a stream of hydrogen or synthesis gas. This vessel was intended to remove oxygen. Any water formed was removed in the condenser and separator (*8*, *12*) followed by the vessels, *M*, containing silica gel.

After passing through the reactor the gases were expanded to atmospheric pressure in the heated valve, *S*; the ammonia was absorbed in the sulfuric acid bubblers, *T*, and the rate of flow of the residual hydrogen-nitrogen was obtained by the wet test meter, *U*.

The reactor itself (Figure 2) was made from a 14-in. length of standard 9/16 × 3/16 in. tubing. 1.625 g. of unreduced catalyst were contained in a 5-cm. length of this tube, and three thermocouples were placed in the bed. In no case was the variation in temperature among these thermocouples more than 3°C., and in most cases it was less than 1°C. The catalyst occupied a space of 0.602 cc., not counting the thermocouple wires (30-gauge iron constantan); the void fraction was 0.46. The particles in the bed were roughly 2 mm. in diameter (9–10 mesh). The catalyst was activated by the standard procedure (*14*). Several external heaters were installed to control the longitudinal temperature variation within the bed.

The design of the reactor described above was determined by the following factors. (1) The large length-to-diameter ratio insured the highest possible linear velocity past the pellets, thus reducing the effect of longitudinal diffusion and diffusion to the catalyst surface to a minimum. (2) The small pellet size reduced the effect of diffusion within the catalyst pores to a minimum. (3) The whole arrangement, with several separately controlled heaters, resulted in very good control of temperature in the bed. (4) The small diameter held the radial temperature differences to minimum values. Usually a bed diameter to pellet diameter of at least ten is used to reduce

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