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February Progress Report

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Attached is the monthly progress report for the Coal Liquefaction Group for February 1980. Included in this report are activities that are going on by CRSD at Linwood in support of the coal liquefaction work. Reports related to all of our contract work with DOE are included here.

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CRDD Coal Liquefaction Group

Monthly Progress Report

February, 1980

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Foam Formation in Wilsonville Recycle Solvent

D. H. S. YING

Summary

Small amount of foam (5% to 8% of fluid volume) was observed in Wilsonville recycle solvent at elevated temperatures (less than 400°F) at high gas input rates. The results indicated the foam formation independent of fluid temperature above 170°F but its stability, i.e. the duration required for the disappearance of foam, reduced continuously with increasing temperature. Gas holdup was found to increase with increasing fluid temperature possibly due to the parallel decrease in fluid viscosity. Furthermore the solvent expanded in a linear rate of 4.8 vol.% per 100°F temperature increase.

Introduction

The gas holdup study with kerosene/nitrogen system, which was reported last month, showed tremendous amount of foam at low gas velocities. Foam formation in this hydrocarbon system initiated the investigation of the possibility of foam formation in SRC process solvent at elevated temperatures. Wilsonville recycle solvent/nitrogen system was used in this work. Due to the constrient of the equipment, the effect of temperature was studied from room temperature to 392°F. In addition, gas holdup and thermal expansion of solvent were investigated.

Equipments and Experimental Method

The equipments consist of a 500 ml graduate cylinder, heating tape, a variable power supply control, a 250°C-ranged thermometer, a gas flow meter, and a stainless steel tube to introduce gas into the solvent. The experiment proceeded by filling half of the cylinder with Wilsonville recycle solvent. Nitrogen gas was introduced at a constant rate. Foam formation and volume expansion were recorded. The gas flow was stopped in order to measure the amount of foam in the case of high turbulence. The stability of the foam was measured simultaneously by recording the duration required for foam disappearance. The final liquid volume was recorded before proceeding to the next gas flow rate. At the end of the ambient temperature study, the fluid was heated to higher temperatures. A constant power supply to the heating tape was used to heat the solvent. The temperature and liquid volume were monitored continuously until steady values were obtained. Then the experiment proceeded with same procedure described above. A total of five temperatures ranging from 65°F to 392°F was used. At the end of the experiments, the volume of the solvent was measured as a function of fluid temperature as it cooled back to room temperature.

Foam Formation

Foam was observed in Wilsonville recycle solvent only at high gas velocities. One of the goals in this work is to determine the existence of foam in SRC-I process solvent at typical high and low gas flow rates at elevated temperatures. It is not intended to determine the transition velocity at which foam is formed. At low gas velocities (approximately 2 cm^3/sec) the solvent did not form foam for the entire temperature range investigated (65°F to 392°F).

At high gas velocities (approximately 150 cm^3/sec.) small amount of foam was observed with a slight dependence on the fluid temperature. Because of the large turbulence at high gas velocities, a distinct layer of foam above the fluid could not be detected. Instead the amount of foam was recorded after shutting off the gas flow, thereby giving the minimum amount of foam formed. By raising the temperature from 65°F (room temperature) to 167°F, the foam increased from less than 2% to 5% of the fluid volume. Further increase in temperature to 392°F did not show substantial change in amount of foam. The stability of the foam decreased continuously with increasing temperature, however. The foam stability is defined as the duration for the foam to completely vanish. As temperature increased, the fluidity of the solvent increased or equivalently the viscosity decreased. It is likely that foam forms more readily in less viscous liquid, explaining the increase of foam in the first increment of temperature raise from 65°F to 167°F. The subsequent insensitive change in the amount of foam with further increase in temperature could possibly be due to the larger uncertainty associated with the amount of foam observed because of the increasing instability of the foam. In any event, the quantity of foam in the Wilsonville recycle solvent (at elevated temperature) is orders of magnitude less than that observed previously in kerosene/nitrogen system. The qualitative aspects of these results were reproducible, such as the effect of temperature on foam stability and amount. However, the absolute quantity of foam formation is not reproducible. One possible explanation is the stripping of the light ends of the solvent during the first set of experiment, thereby reducing the amount of foam when the reproducibility was checked in a subsequent experiment. Nevertheless small amount of foam (5%

to 8%) was observed in Wilsonville recycle solvent at elevated temperatures (less than 400°F) at high gas velocities (0.27 ft/sec. to 0.38 ft/sec. based on the cross section of the graduate cylinder). The results also suggested negligible increase in foam with further increase in temperature above 400°F but the foam stability continuously reduce.

Gas/Hold-Up

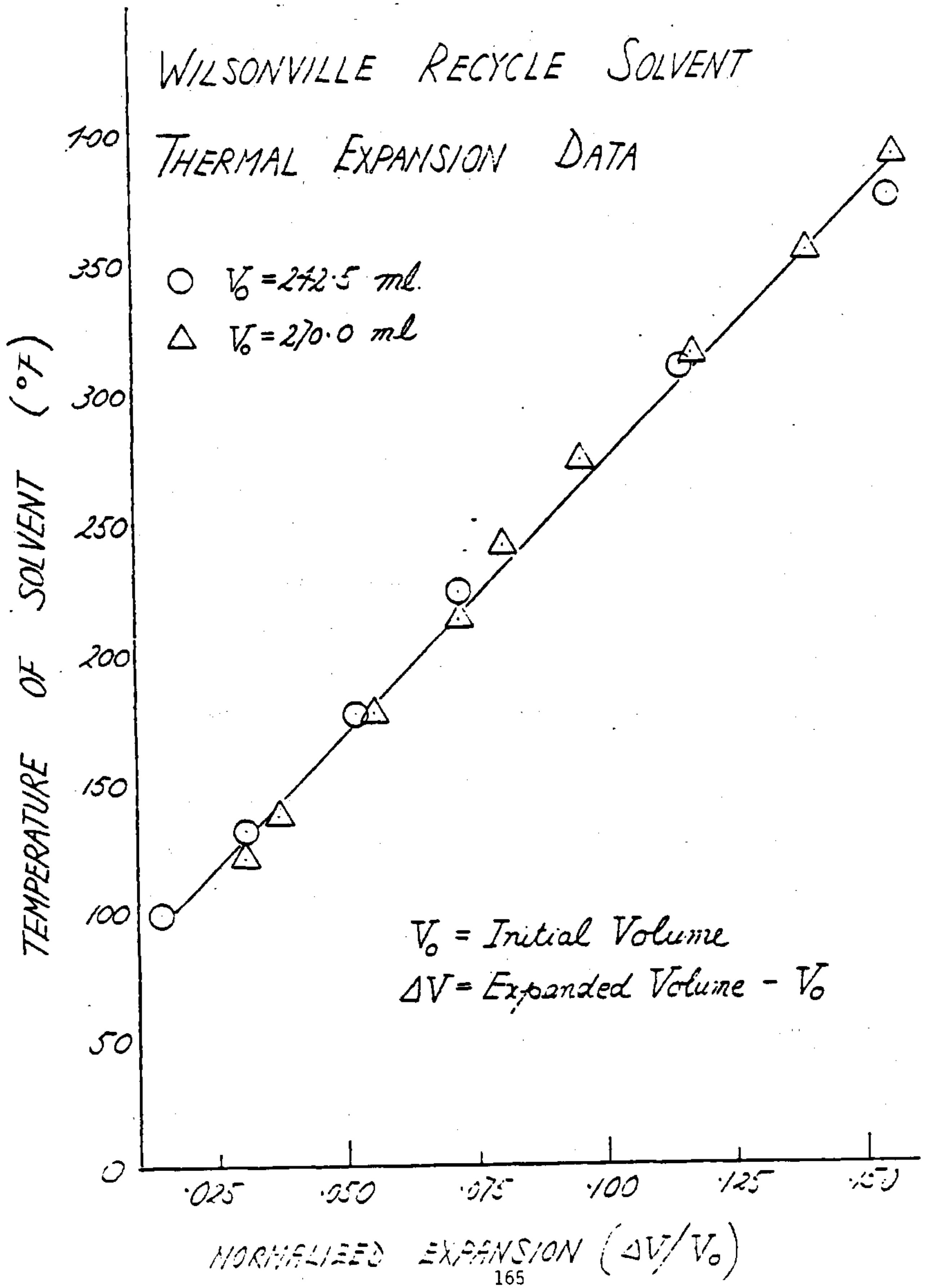
Gas void fraction was found to increase with increasing temperature at a constant gas input rate. The gas void fraction was calculated from dividing the difference between expanded volume and initial volume by the initial ~~expanded~~ volume. The results were summarized in Table (1). Because of the large turbulence at these high gas velocities, the absolute values of the void fraction were not highly reliable. However, a trend of increasing gas holdup with increasing temperature is quite apparent. Although this trend of increasing gas holdup would be partially influenced by foam formation which was unavoidably embedded in the gas void fraction calculation, yet the sizeable difference shown in Table (1) could not be explained by foam formation alone. The increase of gas holdup with increasing temperature could possibly be due to the reduction of fluid viscosity, in agreement with most investigator's work of viscosity effect on gas holdup.

Thermal Expansion of Solvent

Wilsonville recycle solvent expanded considerably with increasing temperature. It was unlikely to accurately monitor the expansion of the solvent during the course of the experiment because gas bubbling stripped part of the solvent

D.Y.

FIGURE (1)



WILSONVILLE RECYCLE SOLVENT
THERMAL EXPANSION DATA
○ V₀ = 242.5 ml.
△ V₀ = 270.0 ml
V₀ = Initial Volume
ΔV = Expanded Volume − V₀
TEMPERATURE OF SOLVENT (°F)
400
350
300
250
200
150
100
50
0
.025
.050
.075
.100
.125
.150
NORMALIZED EXPANSION (ΔV/V₀)

STIRRED REACTOR HYDRODYNAMICS

D. H. S. Ying*

A hydrodynamics study was conducted to gain a further understanding of the characteristics of the unbaffled CPDU (Coal Process Development Unit) stirred reactor in order to improve CPDU data analysis. Specifically, this study included measurements of gas holdup and the dispersion of both gas and liquid phases in a Plexiglas reactor simulator.

Vortex formation, measured in both the air/water and nitrogen/methanol systems, causes the relatively large gas holdup in the unbaffled stirred reactor. Because of this vortex, gas holdup increases dramatically with increasing stirrer speed, but decreases unexpectedly with increasing gas velocity. At typical operating conditions of 40 min residence time, 30 M scf H_2/ton of coal (840°F, 2,000 psig), and 1,000 rpm, gas holdup is estimated to be 18.6%.

The liquid phase is undoubtedly completely back-mixed, as confirmed in a tracer study. Likewise, fine solid particles (silica averaging 10 μm) are homogeneously mixed in the reactor. However, large particles (140-170 mesh glass beads) behave differently; their concentration in the exiting slurry is higher than the average value in the reactor, disagreeing with the completely back-mixed model. This behavior of the large particles can be explained by the solid bypassing phenomenon that results from the centrifugal effect induced by the agitator. Since in the process unit runs the residue particles in the product slurry are very fine (<20 μm), these particles should be well mixed in the CPDU reactor.

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*International Coal Refining Co. (ICRC).

INTRODUCTION

Reaction kinetics and hydrodynamics are two key factors governing the design of the dissolver, a major element in the SRC-I coal liquefaction process. The kinetic data have been obtained in a coal process development unit (CPDU) using a continuous stirred reactor, and the data have been compared with a completely back-mixed model. Although a qualitative observation of gas holdup and solid behavior was performed in the past, this information is not sufficient to fully understand the reactor hydrodynamics. This study was conducted to determine specific fluid dynamic behavior of the continuous stirred reactor to improve the CPDU data analysis. Specific fluid dynamic behavior includes the following:

- Gas holdup
- Liquid dispersion
- Solid dispersion

EXPERIMENTAL PROCEDURE

Fluid dynamic behavior of a stirred reactor depends strongly on the reactor geometry and internal configuration. Two different internal configurations of Plexiglas reactor simulators (Figures 1 and 2) were used in this study. The major differences between them are the width of the agitator blade, the distance of the agitator from the reactor bottom where the inlet is located, and the outlet position. Other minor differences are the dimensions of the thermowell, shaft, and exit opening. Configuration II, depicted in Figure 2, is identical to the CPDU reactor, and most of the experiments were performed with this configuration.

Gas Holdup

Air and water were allowed to flow into the reactor simulator with the agitator rotating at a fixed speed. The air and water exited the reactor through a side opening at the top. After 5 min of steady flow, the operation was ended by simultaneously stopping the agitator, gas flow, and liquid flow. The weight of the liquid retained in the reactor (W_m) was measured. By knowing the weight of liquid occupying the reactor at the same liquid flow rate in the absence of gas and agitation (W_T), the gas holdup or gas volume fraction (ε_g) can be calculated as follows:

$$\varepsilon_g = \frac{W_T - W_m}{W_T}$$

Using liquid weight to calculate gas holdup is more accurate than using liquid volume because the precision in volume measurement is ±2.5 mL, whereas the weight measurement is within ±0.01 g. However, in the gas/slurry three-phase system, slurry volume was used to calculate gas holdup. Air/water, air/water/glass beads (140-170 mesh (105-88 μm)], air/water/silica (average 10 μm), and nitrogen/methanol systems were investigated using configuration II, whereas only the air/water system was tested in configuration I.

Solid Dispersion

Because of the inability of the present system to handle homogeneous slurry feeds, solid dispersion was determined by a special method. Two hundred grams of glass beads or fine silica particles were placed in the configuration II reactor, which was half filled with liquid. Gas and liquid were then passed into the reactor with the agitator operating at a fixed rotating speed. As soon as the slurry began exiting the reactor, a sample was taken. Other samples were collected at fixed intervals of 5, 10, or 20 min. Between each sampling, the exiting slurry was also collected in separate vessels. The solid concentrations in all samples and collecting vessels were calculated by measuring the slurry volume and the weight of the dried solid. The degree of solid dispersion in the reactor was determined by comparing the measured solid concentrations leaving the reactor with predicted values based upon a completely back-mixed model.

Liquid Dispersion

Liquid backmixing in the configuration II reactor was measured by using an approach similar to the solid dispersion study. The conductivity of the liquid leaving the reactor was measured to determine the concentration of a sodium chloride (NaCl) tracer from a calibration curve. The degree of liquid back-mixing in the reactor was determined by comparing these measured concentrations with the completely backmixed model.

RESULTS AND DISCUSSION

Gas Holdup

Configuration I. The effect of stirrer speed on gas holdup is shown in Figure 3, which also provides typical CPDU gas and liquid flow rates. The gas volume fraction increases from 0.04 to 0.20 as the stirrer speed increases from 700 to 1,500 rpm. This five-fold increase in gas holdup with increasing stirrer speed is not surprising because gas holdup is largely due to the vortex formed around the shaft of the agitator. Since the liquid rotates faster as the stirrer speed increases, a deeper vortex is formed, thereby resulting in an increase in the superficial gas holdup. The depth of the vortex increases from 1 to 6.5 in. in the range of stirrer speeds spanned in Figure 3. Because the gas bubbles dispersed around the agitator contribute to a much lesser extent to the total gas holdup, the gas/liquid interfacial area is limited, although the superficial gas holdup is substantial.

Interestingly, gas holdup decreases with increasing gas velocity at constant stirring speed and liquid flow rate, as shown in Figure 4. At both 1,000 and 1,500 rpm, gas holdup is insensitive to the first increment of gas input, but then decreases with further increases in gas velocity. This reduction in gas holdup at higher gas velocities is due to a reduction in the vortex. Experimental results show that vortex depth decreases from 3.75 to 1 in. and from 6.5 to 1.9 in. at 1,000 and 1,500 rpm, respectively, as the gas velocity increases from 180 to 760 cm^3/min. However, as the superficial gas holdup decreases with increasing gas velocity, the amount of dispersed gas bubbles around the agitator (i.e., the gas/liquid interfacial area) increases. Furthermore, this increased amount of dispersed bubbles around the agitator reduces the effectiveness of the agitator to rotate the liquid, thus reducing the vortex depth. At 700 rpm, gas holdup is apparently minimal at all gas velocities, as shown in Figure 4. The same phenomenon of vortex reduction can explain the initial reduction at a gas flow rate of 180 cm^3/min. However, the vortex depth at this rotating speed is initially small (i.e, 1.9 in. at zero gas velocity). Since the measured gas holdup is due to the combined vortex and dispersed bubbles, the subsequent increase in gas holdup at 760 cm^3/min gas flow rate reflects an increased amount of dispersed bubbles because the vortex is smaller. In any event, gas vortex is the major contributor to the gas holdup measured in this system, particularly at high rotating speeds (e.g., 1,000 and 1,500 rpm).

Configuration II. The effect of stirrer speed in configuration II is very similar to that observed in configuration I. Gas holdup is largely dependent upon vortex size, and increases with increasing stirrer speed, as shown in Figure 5. Due to the wider agitator blade in configuration II (i.e., 2 vs. 1.75 in.), the tip speed is higher at the same rotation rate. Consequently, the vortex is larger, and results in a slightly higher gas holdup in the configuration II reactor.

The effect of gas velocity is much less dramatic in configuration II than in configuration I. When gas velocity is increased from 180 to 760 cm^3/min at 1,000 rpm, gas volume percent decreases from 15 to 13.7 (Figure 6), and is also accompanied by a small decrease in vortex size. The smaller reduction in gas holdup with gas velocity in configuration II can perhaps be attributed to the wider agitator blade. The dispersed bubbles around the wider agitator may be less effective at decreasing the liquid rotation speed, which is the key cause of vortex formation. Likewise, at the higher stirrer speed of 1,500 rpm, the vortex size, and consequently gas holdup, are practically unaffected by increasing gas velocity.

In studies of the effect of solid particles on gas holdup, the data fluctuate within only a few tenths of a percent in a wide range of solids concentrations. Therefore, gas holdup is practically unaffected by the presence of solids.

The presence of a vortex in the CPDU reactor precludes the comparison of air/water/gas holdup data with published correlations to predict gas holdup in the actual process, since the literature data were developed in the absence of a vortex. Therefore, a liquid medium having physical properties closely resembling the coal liquid under process conditions should be used to determine gas holdup. The physical properties of methanol are very close to those of coal liquid. As shown in Table 1, the gas holdup in a nitrogen/methanol system at typical operating conditions is higher than that measured in an air/water system (18.6 vs. 15.0%). Since both systems have approximately the same vortex size, the difference in gas holdup reflects the larger amount of dispersed bubbles in methanol, which increases the gas/liquid interfacial area. It should be pointed out that the gas holdup estimate of 18.6% does not account for any foam formation if it exists in the coal system; nevertheless, it is a reasonable value for a nonfoaming system.

Solid Dispersion

The dispersion of solid particles in a stirred reactor was studied with the configuration II reactor. In Figure 7, the concentration of the 140-170 mesh glass beads in the exiting slurry as a function of time was plotted using typical gas and liquid flow rates with the agitator rotating at 1,000 rpm. The results clearly indicate that the measured solid concentration is higher than the expected values based on a completely back-mixed model, represented by the solid line in Figure 7. In the completely back-mixed model, solid concentration in the exiting slurry is identical to the homogeneous concentration in the reactor at any time. This solid concentration is calculated as follows:

$$C = Co \exp (-t/T_R)$$

where C = solid concentration at time t (g/mL), Co = initial solid concentration (g/mL), t = time (min), and T_R = slurry residence time (min).

Solid bypassing, depicted in Figure 8, is one possible explanation for the higher solid concentration in the exiting slurry. Liquid rotation induced by the agitator acts like a centrifuge, throwing the solid particles toward the wall of the reactor, and resulting in a radial solid concentration gradient. Since the exit line is located at the side, the higher concentration slurry will preferentially exit the system, resulting in the data in Figure 7.

The degree of solid bypassing decreases with increasing gas velocity. Figure 9 plots the concentration of glass beads in the exiting slurry as a function of time and increasing gas velocity. During the first 30 min, the slurry leaving the reactor still has a higher solids concentration than the back-mixed model would predict, but the magnitude is less than that measured at lower gas velocity (Figure 7). This effect of gas velocity can also be explained by the solid bypassing concept. Similar to the effect of gas velocity on gas holdup, the increased amount of dispersed bubbles around the agitator as a result of increasing gas velocity reduces the effect of liquid rotation. Consequently, the centrifugal influence is decreased, and yields a lower radial concentration gradient. Therefore, the measured exiting solid concentration is lower at higher gas velocity.

If the centrifugal effect is the major cause of the observed higher exiting solid concentration, one would expect the influence of centrifugal force to decrease by reducing the particle size. A similar plot of the solid concentration versus time for very fine silica particles averaging 10 μm at typical operating conditions is depicted in Figure 10. The results show good agreement with the back-mixed model, which further supports the solids bypassing concept, and suggests that the effect of centrifugal force in this fine-particle system is only moderate.

Since the feed coal in the CPDU is 150 mesh and finer, the median coal particle size is small. Therefore, the 140-170 mesh glass bead results represent the worst possible case, which is very unlikely to occur. In addition, as the coal dissolves, the particle size is significantly reduced. The residual material in the product slurry consists of particles less than 20 μm. Therefore, based upon the results of this solid dispersion study, the solid particles in the CPDU stirred reactor are speculated to be homogeneously mixed.

Liquid Dispersion

The degree of liquid back-mixing in the stirred reactor was examined under typical flow conditions. The exiting tracer concentration as a function of time is plotted in Figure 11. The good agreement between the data and the back-mixed model prediction indicates a completely well-mixed liquid phase in the reactor. This agreement also supports the data on the well-mixed fine solid particles shown in Figure 10, thereby indicating a homogeneous slurry mixture in the continuous stirred reactor.

CONCLUSIONS

Gas holdup in the stirred reactor was found to depend strongly on stirrer speed and slightly on agitator size. A five-fold increase in gas holdup was observed by doubling the stirrer speed. This strong dependence on stirrer speed was primarily due to vortex formation in the unbaffled reactor. More interesting is the reduction in gas holdup with increasing gas velocity. At typical operating conditions in the CPDU, gas holdup is 18.6%.

Vortex size was found to decrease with increasing gas velocity. One explanation is the increased amount of dispersed bubbles around the agitator with increasing gas velocity, which reduces the effectiveness of liquid rotation. Consequently, the vortex is reduced accordingly with the decrease in liquid rotation.

Undoubtedly, the liquid phase is completely back-mixed. Since fine silica particles were found to be homogeneously mixed in the reactor, and the residue particles in the product slurry are in the same size range (<20 μm), it is believed that the solid phase is also completely back-mixed. However, the other effects influencing solid dispersion, such as solid density and rate of coal dissolution, should also be considered.

This short study has provided an understanding of the behavior of the CPDU stirred reactors. However, more detailed information is needed, including the effect of solid density on solid dispersion, the mechanisms eliminating vortex (baffle has been tried and causes coke formation), the effect of foaming on gas holdup, and gas/liquid mass transfer rate.

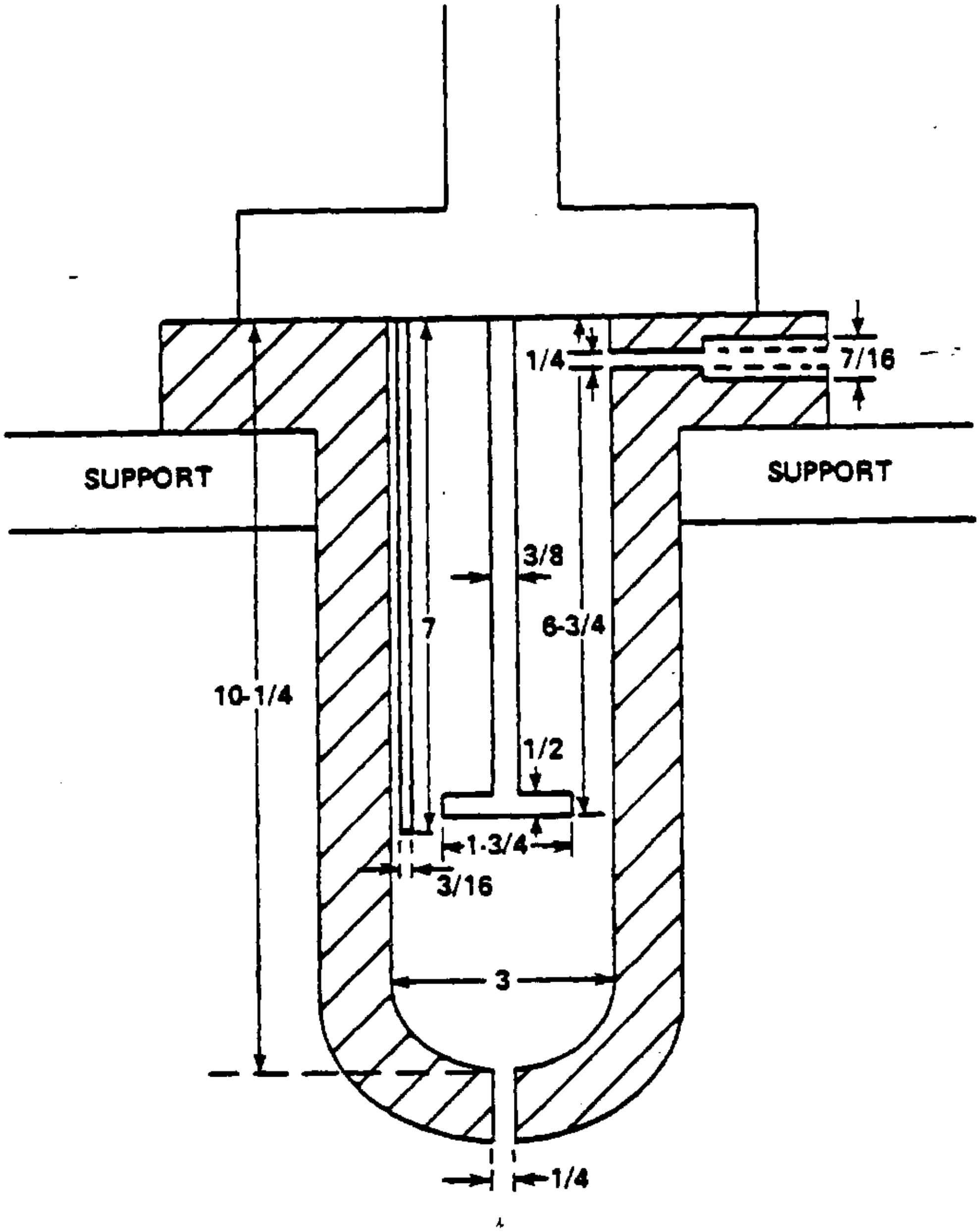
Table 1

Gas Holdup in Two Mixture Systems[a]

	Air/water	N_2/methanol
Gas holdup (%)	15.0	18.6
Liquid properties		
Density (g/mL)	1.00	0.79
Viscosity (cP)	1.00	0.58
Surface tension (dyne/cm)	72.8	22.3

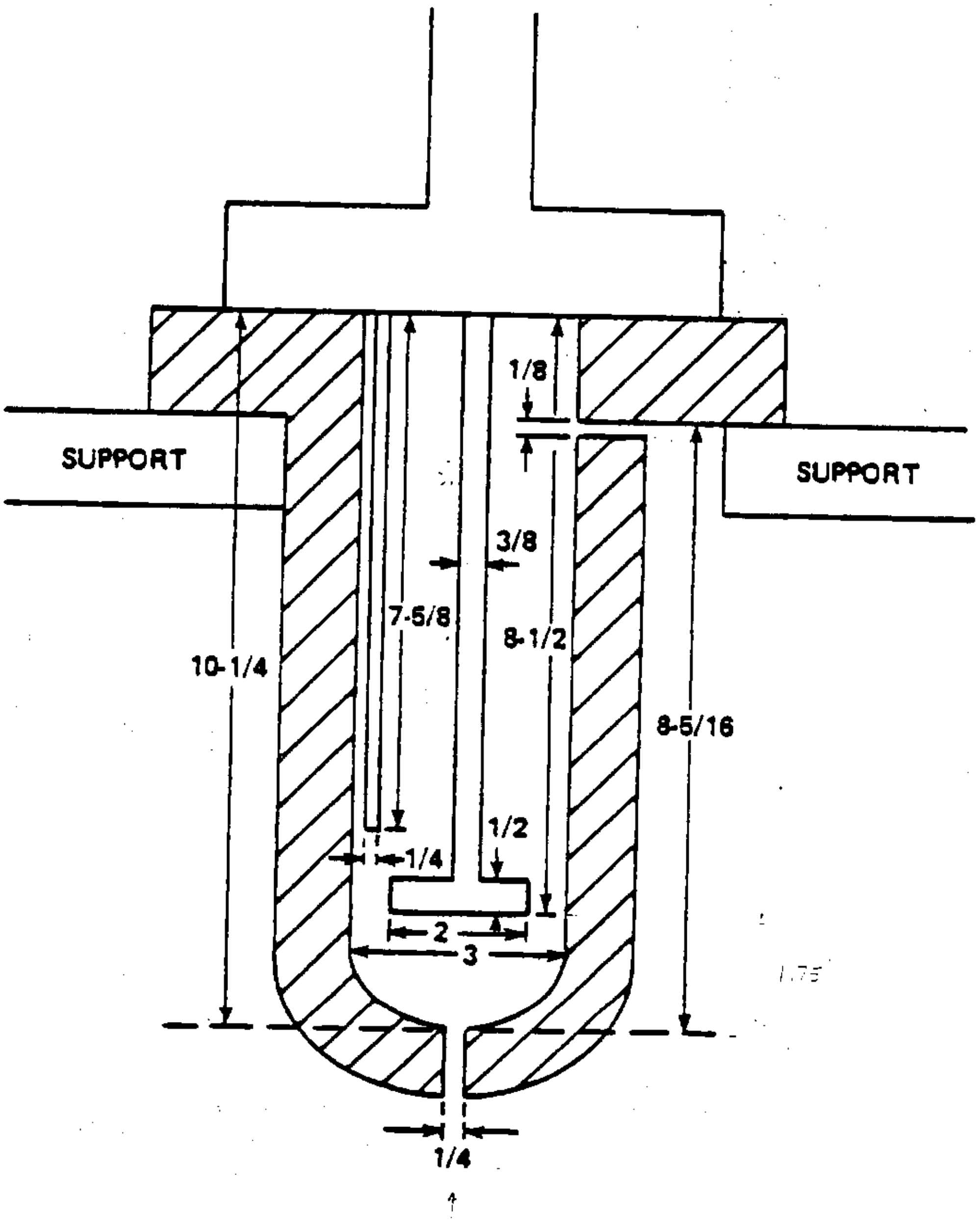
[a] Configuration II; 1,000 rpm; gas flow rate = 180 cm^3/min; liquid flow rate = zero and 25 cm^3/min.

Figure 1
Schematic of Configuration I Reactor*



*ALL DIMENSIONS ARE IN INCHES

Figure 2
Schematic of Configuration II Reactor*



*ALL DIMENSIONS ARE IN INCHES

Figure 3
Effect of Stirrer Speed on Gas Holdup
Air/Water System

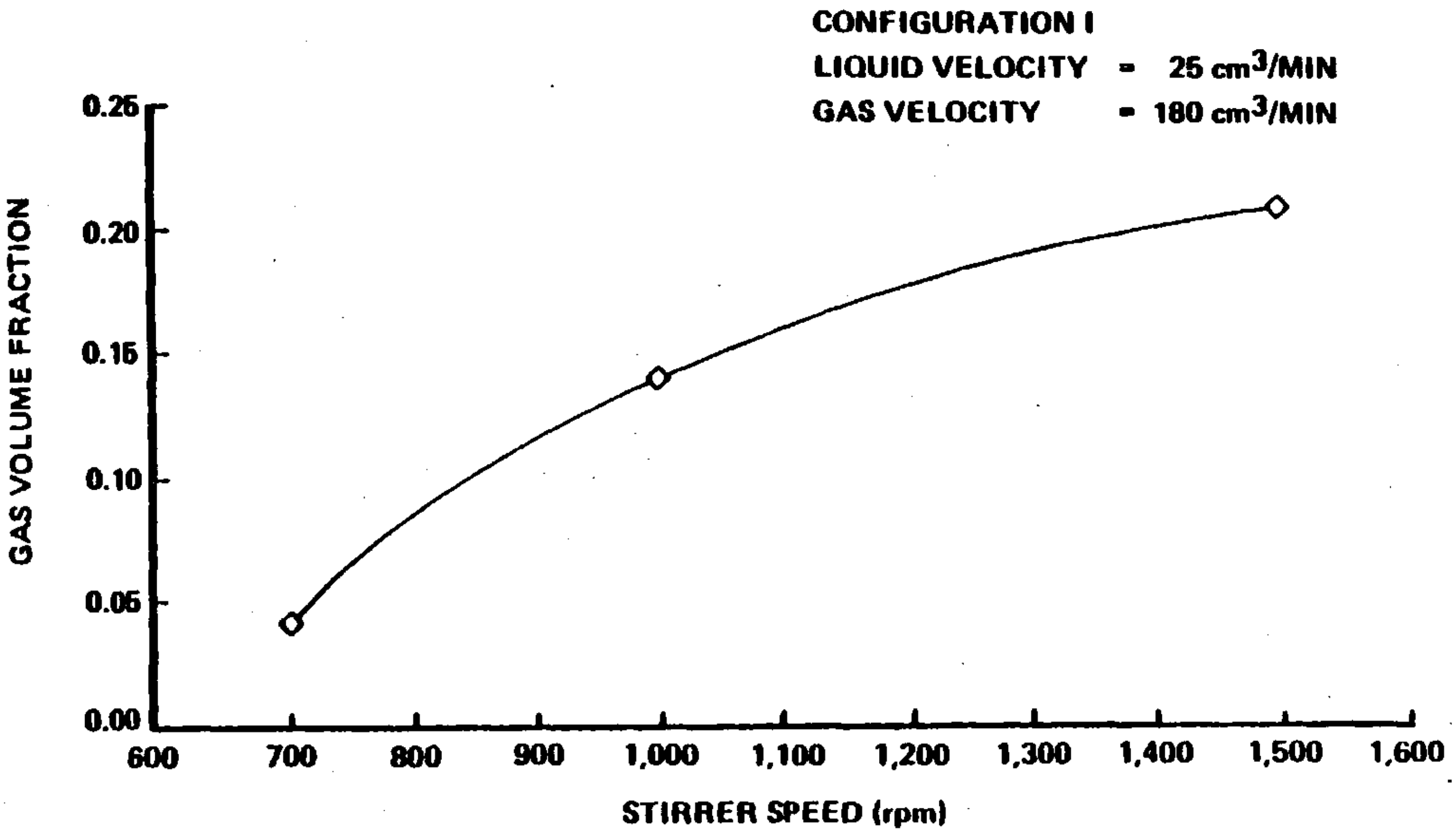


Figure 4
Effect of Gas Velocity on Gas Holdup
Air/Water System

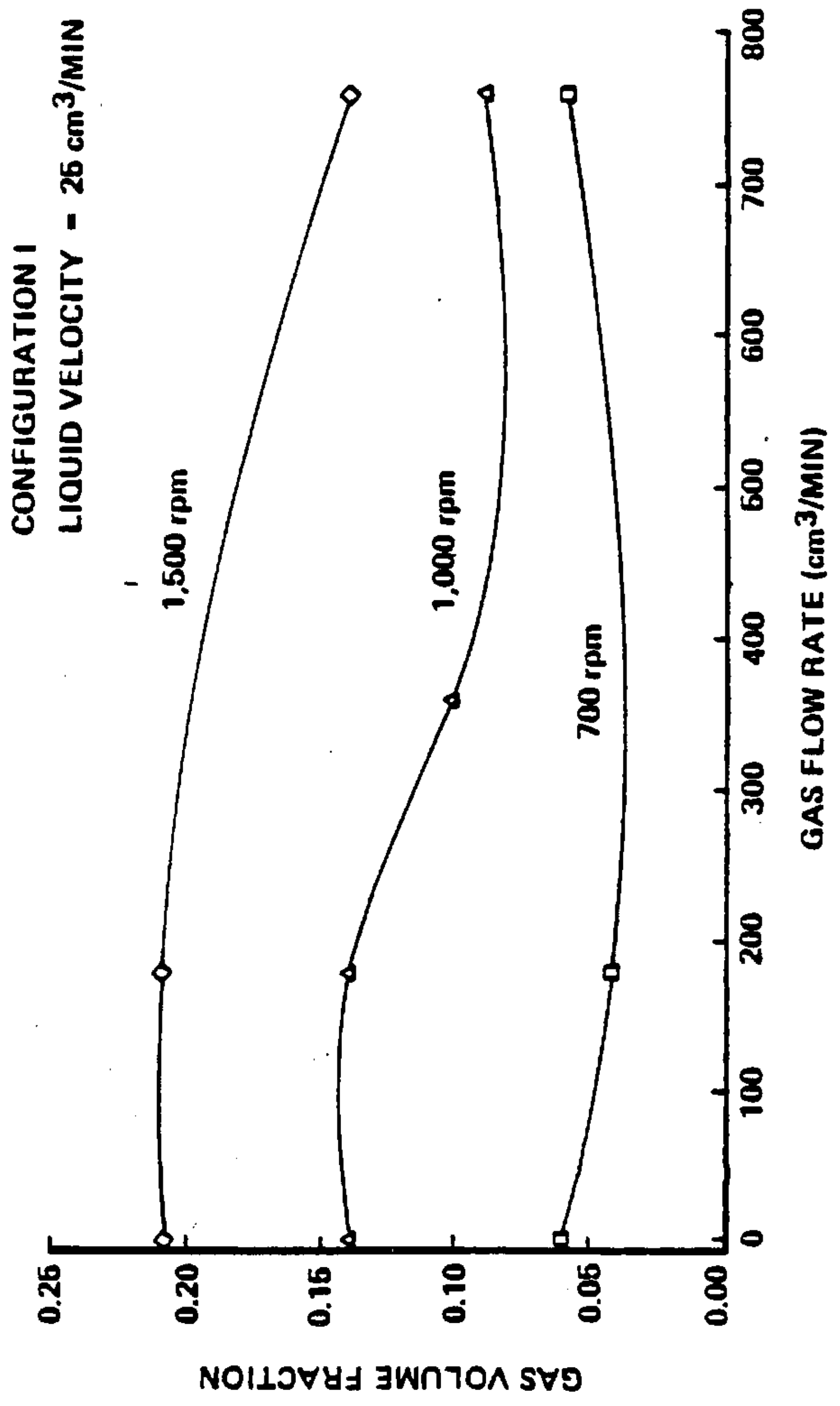


Figure 6
Effect of Stirrer Speed on Gas Holdup
Air/Water System

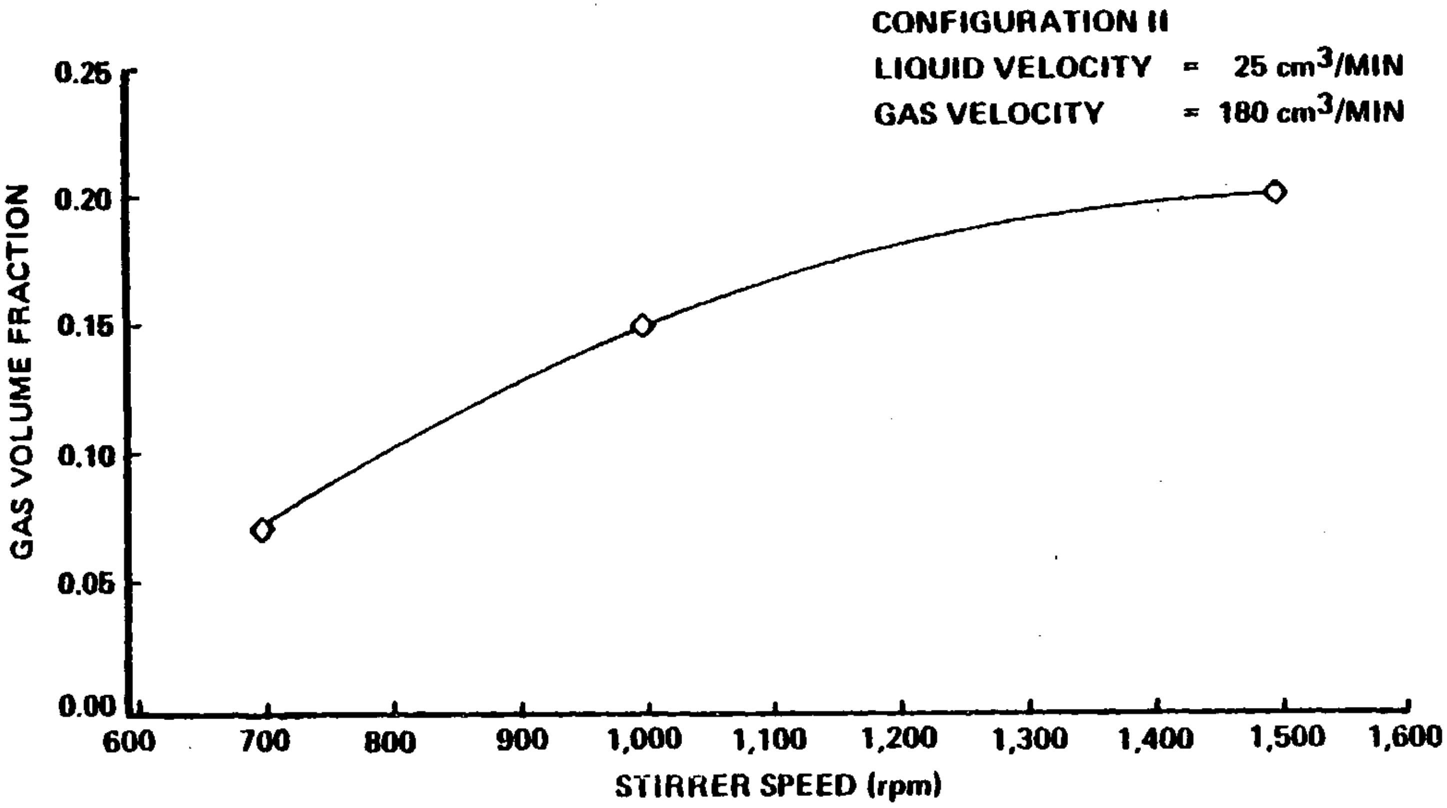


Figure 6
Effect of Gas Velocity on Gas Holdup
Air/Water System

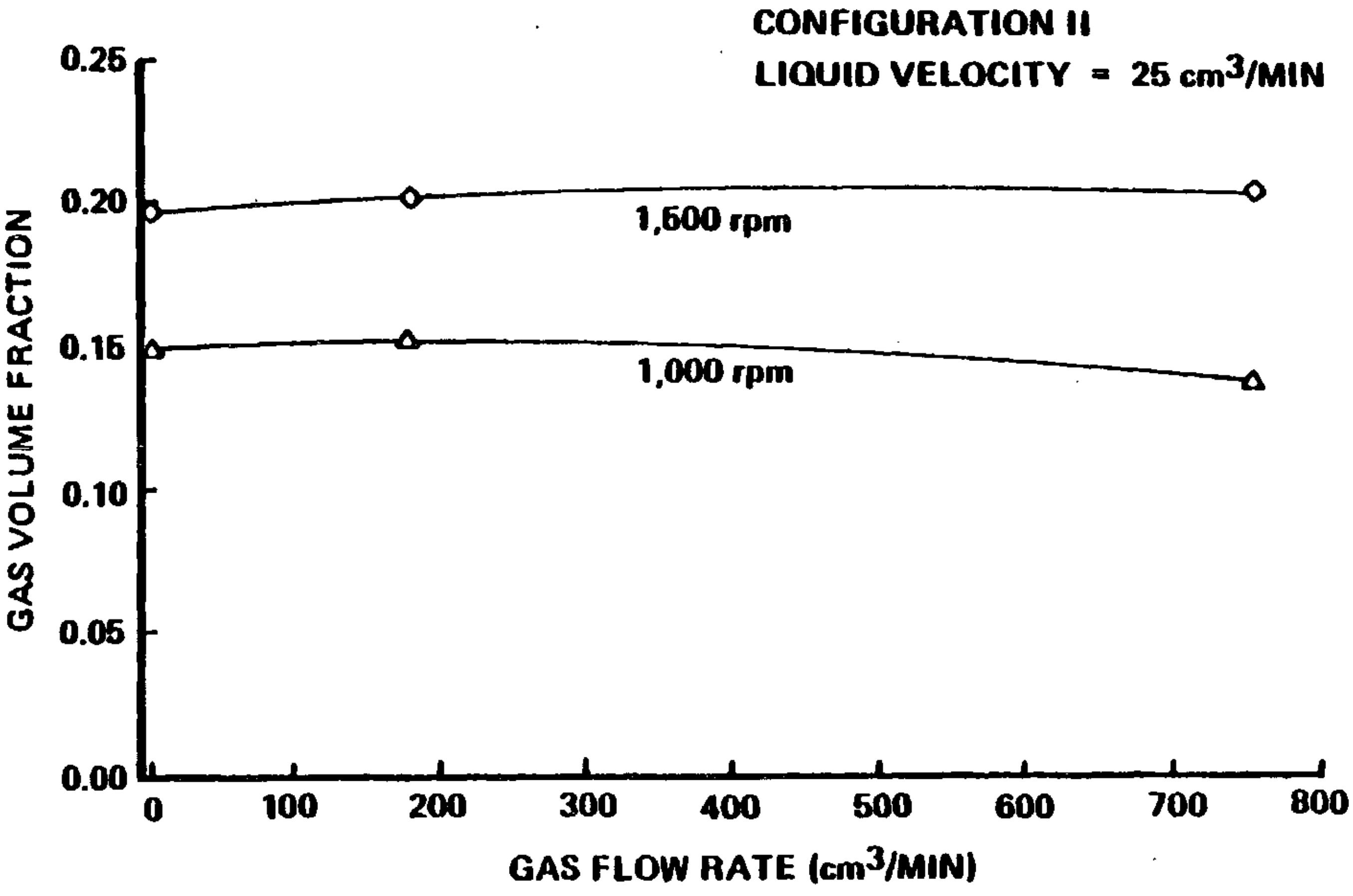


Figure 7
Solid Dispersion
Air/Water/Glass-Bead System

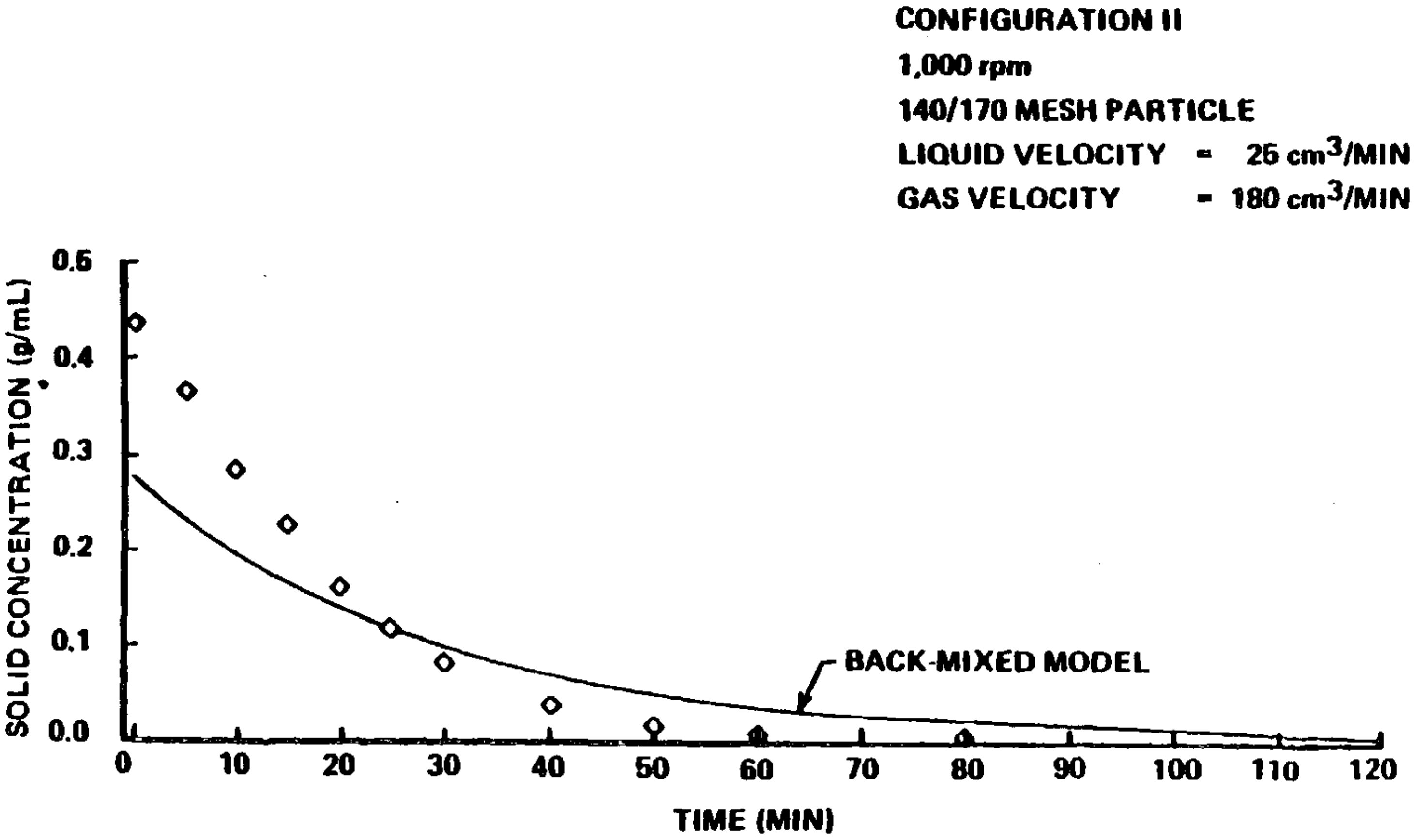


Figure 8
Depiction of Solid Bypassing Concept

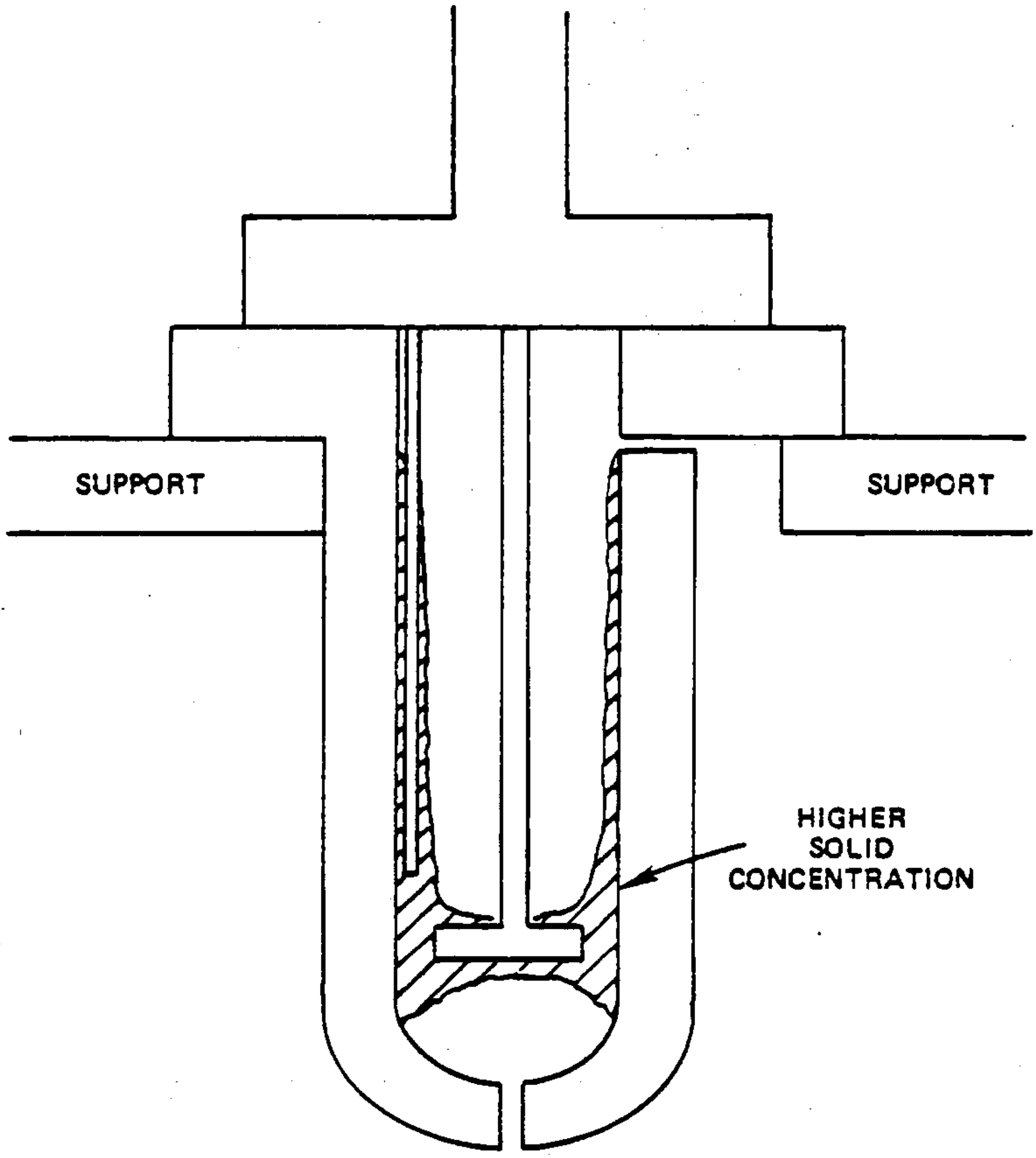
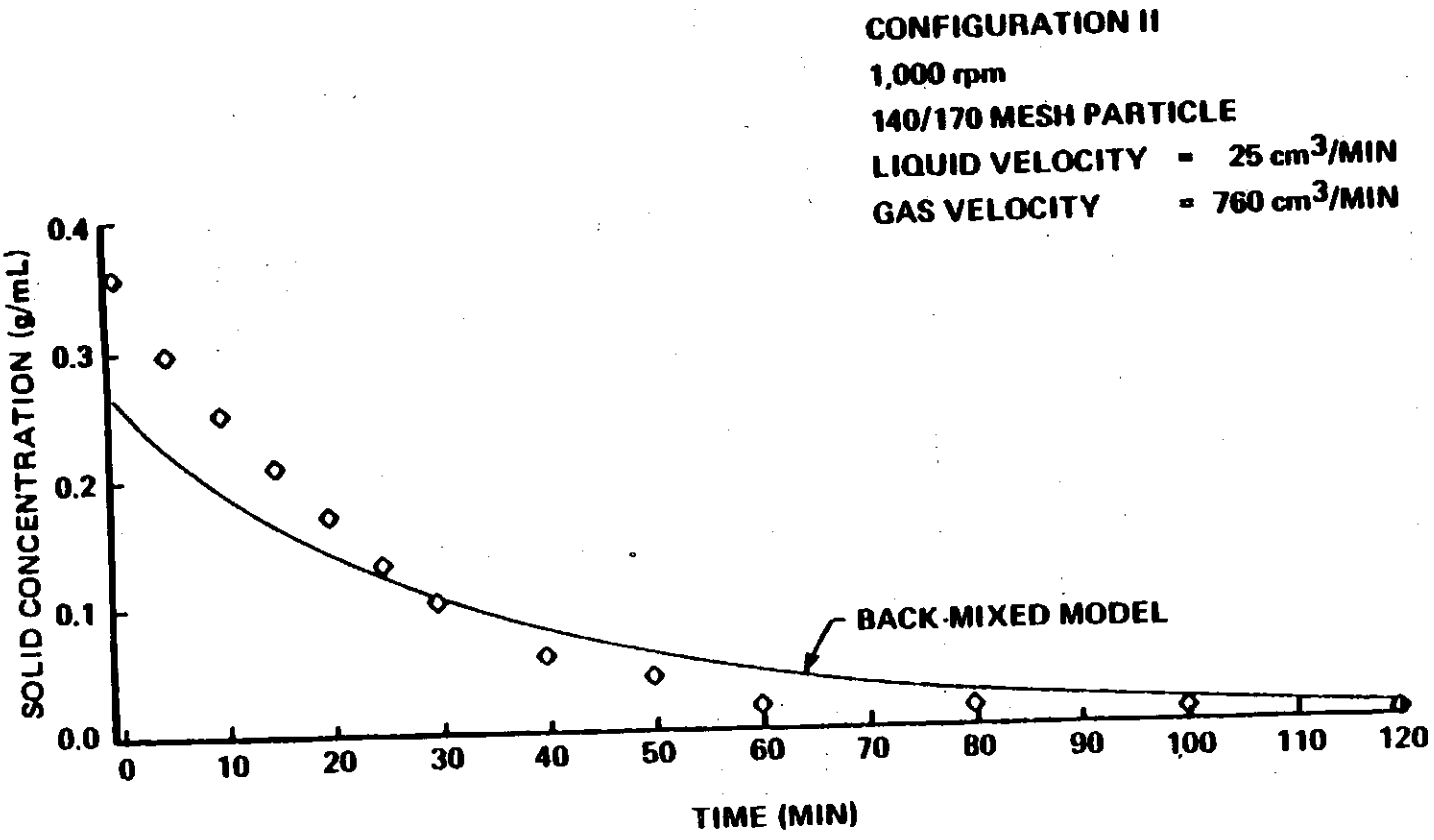


Figure 9
Solid Dispersion
Air/Water/Glass-Bead System



CONFIGURATION II
1,000 rpm
140/170 MESH PARTICLE
LIQUID VELOCITY = 25 cm^3/MIN
GAS VELOCITY = 760 cm^3/MIN

Figure 10
Solid Dispersion
Air/Water/Silica System

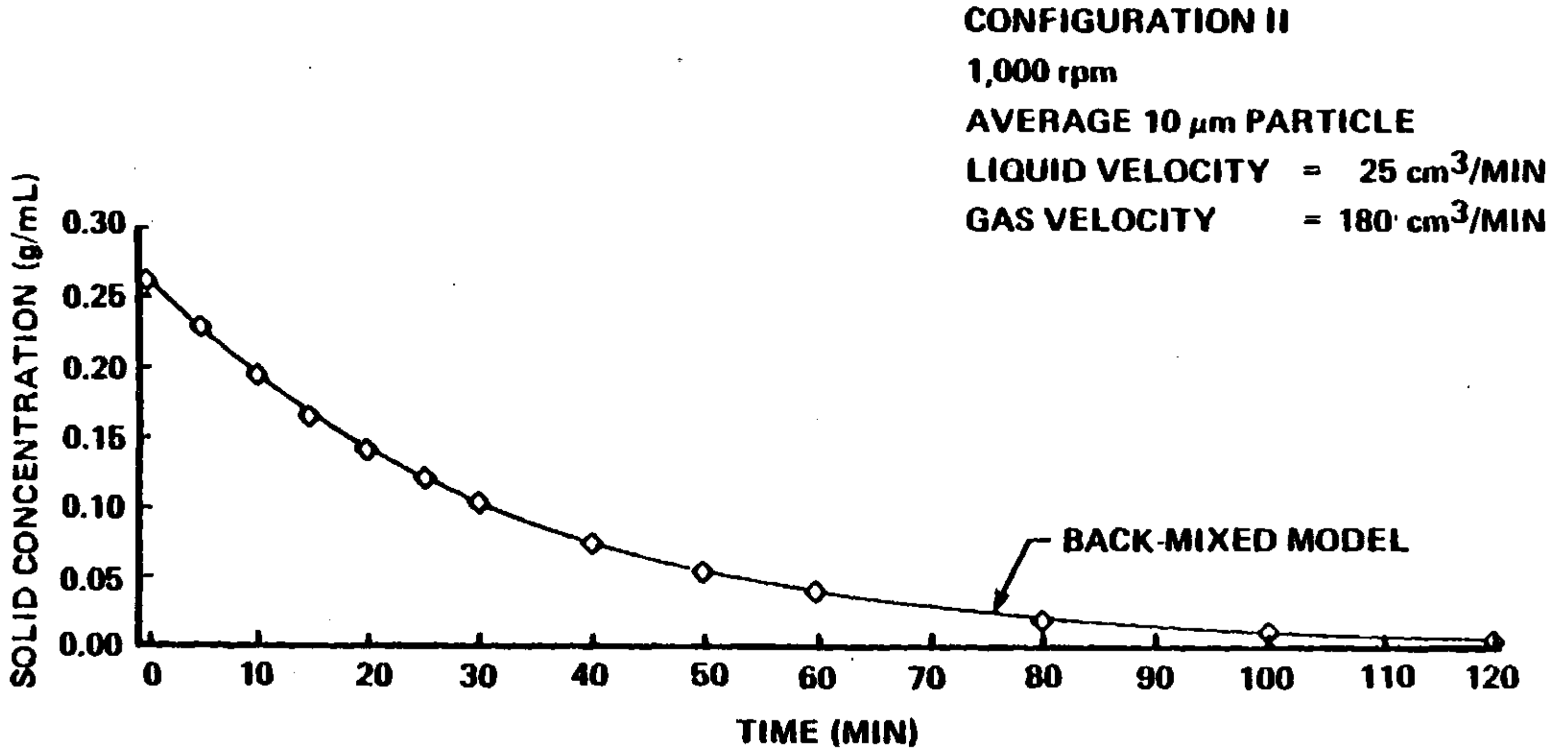
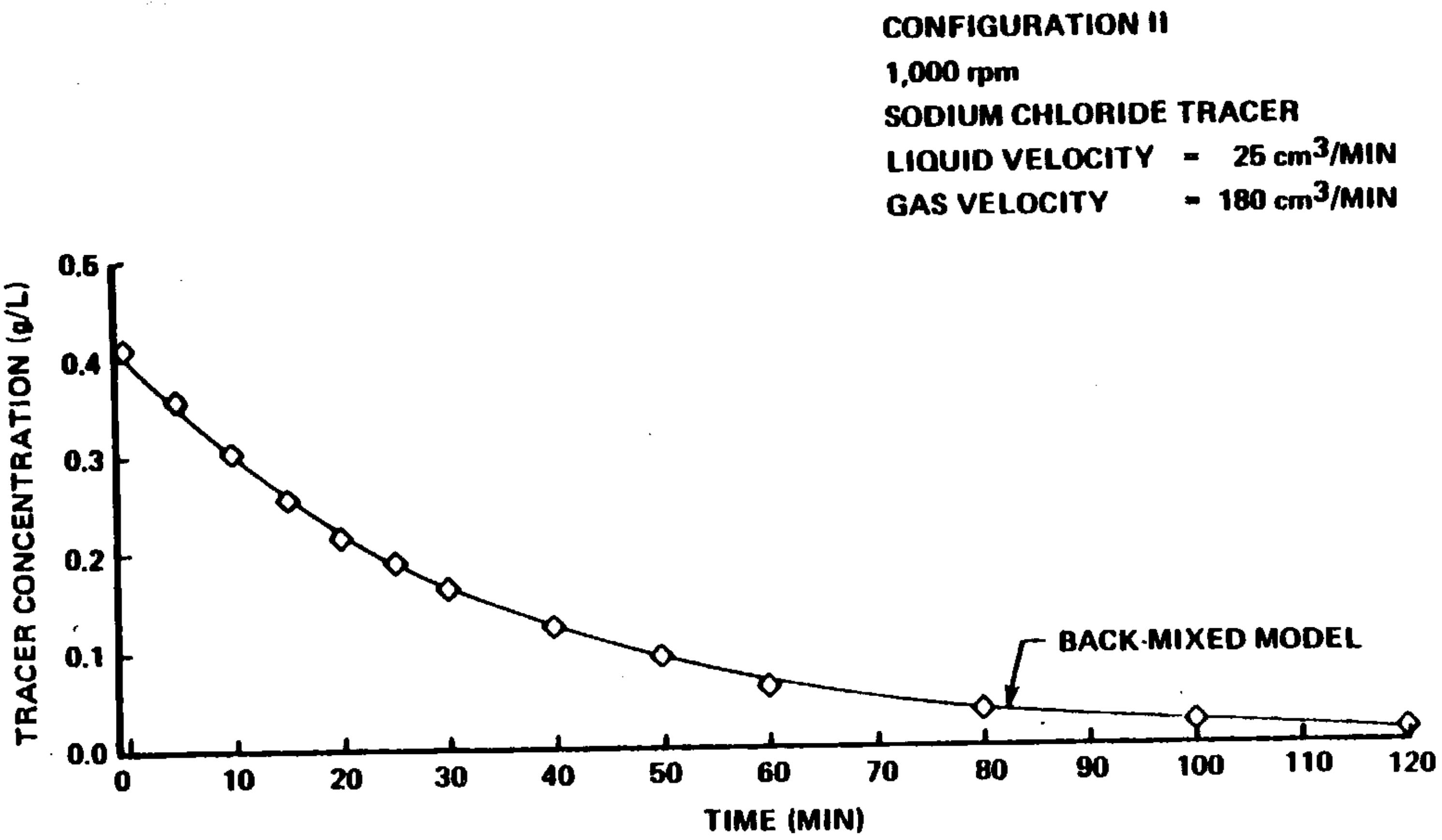


Figure 11
Liquid Dispersion
Air/Water System



Gas Holdup in Gas-Liquid and Gas-Liquid-Solid Flow Reactors

D. H. S. Ying, E. N. Givens, and R. F. Weimer

Air Products and Chemicals, Inc.
Allentown, Pennsylvania

Abstract

A recent article by Lee, Guin and Tarrer raised a number of questions concerning the methodology for designing a three-phase reactor for the solvent refining of coal (SRC). Laboratory studies of two- and three-phase flow in subscale models of an SRC reactor have now resolved several of these questions. In particular, water/nitrogen and methanol/nitrogen gas holdup data from our 2-inch and 5-inch-diameter columns are in good agreement with the correlation of Yoshida and Akita. The presence of silica particles (140 mesh minus and 30-45 mesh) reduces gas void fraction at low superficial gas velocities. However, at higher gas flow rates (0.20 ft/sec) the reduction effect of solid particles diminishes to a negligible level, suggesting that the correlation of Yoshida and Akita is equally adequate for a gas-liquid-solid system at high gas superficial velocities. Furthermore, gas distribution and liquid flow rate show no effect on gas holdup.

Introduction

Gas holdup in bubble columns has been a subject of great interest, particularly in reactor column design. Numerous investigators have studied gas holdup with many different systems, covering a wide range of liquid viscosity (0.58-152.0 cp), surface tension (22.3-76.0 dynes/cm), and density (0.79-1.70 gm/cc). Recently Tarrer et al. (1978) discussed the evaluation of gas holdup in bubble columns in conjunction with a dispersion model for the solvent refined coal process. In particular, the authors examined the general applicability of two gas holdup correlations. One was that of Hikita and Kikukawa (1974) who proposed the expression

$$\epsilon_g = 0.505\, V_g^{0.47} \, (72/\sigma)^{2/3} \, (1/\mu)^{0.05} \qquad (1)$$

The other correlation was from Yoshida and Akita (1973) who expressed the gas void fraction as a complex function of three dimensionless functional groups,

$$\frac{\epsilon_g}{(1 - \epsilon_g)^4} = C_1 \, (N_{Bo})^{1/8} \, (N_{Ga})^{1/12} \, (N_{Fr}) \qquad (2)$$

where

$$N_{Bo} = gD^2 \rho_L / \sigma \qquad \text{(Bond number)} \qquad (3)$$

$$N_{Ga} = gD^3 / \nu_L^{\,2} \qquad \text{(Galileo number)} \qquad (4)$$

$$N_{Fr} = V_g / \sqrt{gD} \qquad \text{(Froude number), and} \qquad (5)$$

C_1 = 0.20 for nonelectrolyte solution, 0.25 for electrolyte solution

Equation (2) also predicts a holdup which is independent of column diameter, and it may be written in terms of only two dimensionless groups, such as

$$\frac{\epsilon_g}{(1 - \epsilon_g)^4} = C_1 \left(\frac{\rho_L^3 \nu_L^4 g}{\sigma^3} \right)^{1/24} \left(\frac{V_g^3}{g \nu_L} \right)^{1/3} \qquad (6)$$

Tarrer et al. arrived at the general conclusion that Equation (2) should be used when column diameter (D) is greater than 5.9 in (15 cm), while an average of Equations (1) and (2) was recommended for design purposes when D is less than 5.9 in.

Air Products and Chemicals has been involved in the detailed design of a 6000 T/D demonstration plant for the solvent refining of coal through a DOE contract. As presently envisioned, the dissolver in the demonstration plant will be 11 feet in diameter and 110 feet tall. As part of this design effort, gas holdup in gas-liquid and gas-liquid-solid flow reactors was investigated.

Recent work in our laboratory, using water-nitrogen and methanol-nitrogen in 2-inch diameter (5.08 cm) and 5-inch diameter (12.7 cm) Plexiglas columns, has demonstrated good agreement with the correlation [Equation (2)] of Yoshida and Akita. In all cases, the correlation of Hikita and Kikukawa [Equation (1)] overestimates the gas holdup. The average of these two correlations does not fit our results as well as Yoshida's expression alone. We have also found that neither the gas distributor nor the liquid flow rate affect gas holdup. An important feature of the present work was the measurement of gas holdup both in the presence and the absence of suspended solids. The data show that the gas holdup is reduced by the presence of suspended solid particles at low superficial velocities. However, at higher superficial velocities, there is very little difference between the two holdups. This almost identical gas holdup suggests adequate application of the correlation of Yoshida and Akita for gas-liquid-solid system at high gas flow rates. Moreover, the effect of solids appears to be stronger in a five-inch diameter column than in a two-inch diameter column. At relatively low superficial gas velocities, ignoring the effect of solid particles on gas holdup might lead to significant error. In addition, a 1-foot diameter by 25 feet tall glass column has been constructed at Air Products and Chemicals to study fluid flow parameters, including gas holdup, for both gas-liquid and gas-liquid-solid systems. Results from this large column will be coupled with the present work to provide better confidence for scaling up to the demonstration dissolver.

Experimental

Gas holdup studies were performed in 2-inch diameter (5.08 cm) and 5-inch diameter (12.7 cm) Plexiglas columns which are both 5 feet (1.52 m) high. The experiments were performed in the absence of liquid flow by completely filling the column with liquid (water or methanol) and then passing nitrogen through the liquid at specified rates. Excess liquid exited the column at the top through a side opening. A waiting period of 10 to 15 minutes was allowed to ensure steady state condition achieved. The bottom valve was then closed to shut off the gas input. The final liquid level was measured, and the difference between the initial (top exit opening) and the final levels represented the gas holdup at that particular gas flow rate. Measurements were also made with a gas distributor plate with seven 0.25-inch (0.64 cm) openings inserted into the 5-inch diameter column.

Both 30-45 mesh (600 μm-350 μm) and 140 mesh minus (less than 100 μm) silica particles were employed to investigate the effect of solids suspension on gas holdup. To ensure a true solid suspension effect a slightly modified technique was used. At the start of an experiment the top exit opening was valved off as gas passed through the column to allow complete solid suspension to develop. Then the valve was opened to empty any excess solution. The rest of the experiment proceeded as described above.

Continuous silica-water slurry flow experiments were also conducted. The slurry and nitrogen passed into the column through a centrally located opening at the bottom. After steady state was reached, a common valve at the bottom was closed stopping both slurry and gas flow simultaneously. The gas void fraction was measured as described above.

Results and Discussion

Gas holdup data in the water/nitrogen system are shown in Figure (1). Excellent agreement was obtained between Yoshida's correlation and the results from the 5-inch diameter column. The results from the 2-inch diameter column also show good agreement at low gas input rates, but significant deviations were observed at superficial velocities above 0.10 ft/sec (3.05 cm/sec). At this gas input rate, fully developed slug flow was observed in the 2-inch diameter column. In the larger column (5-inch diameter) slug flow behavior was not seen up to the maximum superficial velocity of 0.27 ft/sec (8.23 cm/sec) attainable with the equipment.

The gas void fraction predicted by the correlation of Hikita and Kikukawa [Equation (1)] was always higher than the experimental results from both columns at low superficial gas velocities, as shown in Figure (1). The prediction from Equation (1) apparently approaches our gas holdup results (5-inch diameter column) at higher gas superficial velocities. However, as mentioned by Tarrer, et al., the application of Equation (1) becomes limited at high gas superficial velocities, since it predicts a gas void fraction greater than unity.

The methanol/nitrogen system showed similar behavior as shown in Figure (2). The average bubble size is smaller in methanol than in water because of the lower surface tension of methanol, resulting in a slightly larger gas holdup. The deviation between the gas holdup measured from the 2-inch diameter and 5-inch diameter columns is less in the methanol/nitrogen system, presumably because the wall proximity effect is lower for small bubbles than for larger ones. Yoshida's correlation shows reasonably good agreement with the methanol/nitrogen gas holdup results. However, the prediction from Equation (1) is much too high. Since the small dependence on the liquid viscosity shown in Equation (1) has negligible impact on the gas holdup, the dependence on the liquid surface tension must be responsible for this high gas holdup prediction. The correlation of Yoshida and Akita, on the other hand, appears to handle the effect of surface tension very well, since it is very close to the experimental data for both water/nitrogen and methanol/nitrogen systems. This suggests the general application of Yoshida's correlation for low-surface-tension systems. In addition, the correlation of Yoshida and Akita correctly predicts that at low gas flow rates, the bubble rise velocity is independent of superficial gas velocity. This is more obvious when Equation (6) is rearranged to give an explicit expression for the bubble rise velocity (V_{BR}) as $\epsilon_g \ll 1$.

$$\frac{V_{BR}}{(g \nu_L)^{1/3}} = \frac{1}{C_1} \left(\frac{\sigma^3}{\rho_L^3 \nu_L^4 g} \right)^{1/24} \qquad (7)$$

where $V_{BR} = V_g / \epsilon_g$

Substitution of the physical properties of water gives V_{BR} a value of 1 ft/sec, which approximates the terminal velocity of single bubbles in water [Datta, et al. (1950)].

Tarrer, et al. has recommended an average of Equations (1) and (2) for design purposes when the column diameter is less than 5.9 in (15 cm). However we found that an average of these two equations does not fit our results from the 5-inch-diameter column as well as Equation (2) alone. In fact the correlation of Hikita and Kikukawa is not dimensionless, making it difficult for scale-up of bench-scale data.

When a distributor plate with seven 0.25-inch openings was used in the 5-inch diameter column, the measured gas holdup in water was identical to that obtained without the plate at gas velocities up to 0.09 ft/sec (2.74 cm/sec). At higher gas input rates, gas was trapped below the distributor plate. This extra gas pocket displaced more fluid, causing an apparently larger gas void fraction. After correcting for this gas pocket effect, the gas void fractions at high gas input rates, as shown in Figure (3), agreed with those without the distributor plate (Figure (1)). This agreement shows that the presence of a distributor plate does not affect gas holdup in the section of the bubble column above the plate.

The presence of suspended solid particles in a bubble column reduces the gas holdup. This effect is somewhat dependent on the column diameter. The comparison of the gas holdup in a 2-inch diameter column with and without suspended solid (140 mesh minus) shows a negligible difference, as shown in Figure (4). When the column diameter was increased to 5 inches, less gas holdup was observed in the presence of the suspended solids (140 mesh minus) as shown in Figure (5). A gas holdup study with air-water-glass spheres by Kato et al. (1972), showed similar results. They reported visual observation of higher rising velocity of the coalesced bubbles in the presence of solid particles. In our system the suspended silica particles made the slurry column opaque, and visual observation of bubble size and rising velocity was not possible. In any event, the difference in gas holdup is less obvious at high gas flow rates. This also agrees with the observation of Kato et al. (1972), and it suggests that high gas turbulence reduces the coalescence effect caused by the presence of suspended solid. An identical reduction effect at low superficial gas velocities was observed when larger particles (30-45 mesh) were used in the 5-inch diameter column. With these larger particles large coalesced bubbles were observed and the reduction effect extended to slightly higher gas input rates (0.19 ft/sec versus 0.13 ft/sec) as shown in Figure (6). This suggests that the gas turbulence required to reduce the coalescence effect is higher for larger solid particles. Increasing the average solid concentration from 15.7 wt % to 45.0 wt % produced no further systematic change in the gas holdup as shown by Figure (5).

Our laboratory data showed that gas holdup in bubble columns with suspended solid particles was not affected by the slurry flow rate. Three different slurry input rates, ranging from 5 cc/sec to 24 cc/sec, were employed in testing the slurry flow rate effect on gas holdup in the 5-inch diameter column. The results showed negligible differences between the gas holdups measured with and without slurry flow.

Both in the presence and absence of suspended solid particles, gas holdup at low gas velocities is by itself a small quantity. For instance, at a superficial gas velocity of 0.06 ft/sec (corresponding roughly to the gas feed rate in the Wilsonville SRC pilot plant), the gas void fraction was reduced from [illegible] to 0.04 in the presence of solid particles as shown by Figure (5). This reduction has little effect on the liquid holdup. However, it is important to note that this change represents a 20% reduction in the gas void volume, which together with changes in bubble size could appreciably affect mass transfer rate. Consideration of the effect of solids suspension on bubble behavior may be significant in the design of three-phase slurry reactors.

Nomenclature

ϵ_g	gas void fraction
V_g	superficial gas velocity [m/sec in Equation (1); cm/sec in Equations (5) and (6)]
σ	surface tension of liquid (dyne/cm)
μ	viscosity of liquid (cp)
g	gravitational constant (cm/sec^2)
D	diameter of column (cm)
ρ_L	density of liquid (gm/cm^3)
ν_L	kinematic viscosity of liquid (cm^2/sec)
V_{BR}	bubble rise velocity (cm/sec)

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Figure (1) Gas Holdup in Water/Nitrogen System

□ 2-inch-diameter column
○ 5-inch-diameter column

Figure (2) Gas Holdup in Methanol/Nitrogen System

□ 2-inch-diameter column
○ 5-inch-diameter column

Figure (3) Effect of Distributor Plate on Gas Holup in a 5-inch-diameter Column

Figure (4) Effect of Solids on Gas Holup in a 2-inch-diameter Column

Figure (5) Effect of Solid Concentration on Gas Holup in a 5-inch-diameter Column

Figure (6) Effect of Large Solid Particles (30-45 mesh) on Gas Holdup in a 5-inch-diameter Column

Figure (1)

Gas Holdup in Water/Nitrogen System

- ° 2-inch-diameter column
- ° 5-inch-diameter column

WATER/NITROGEN

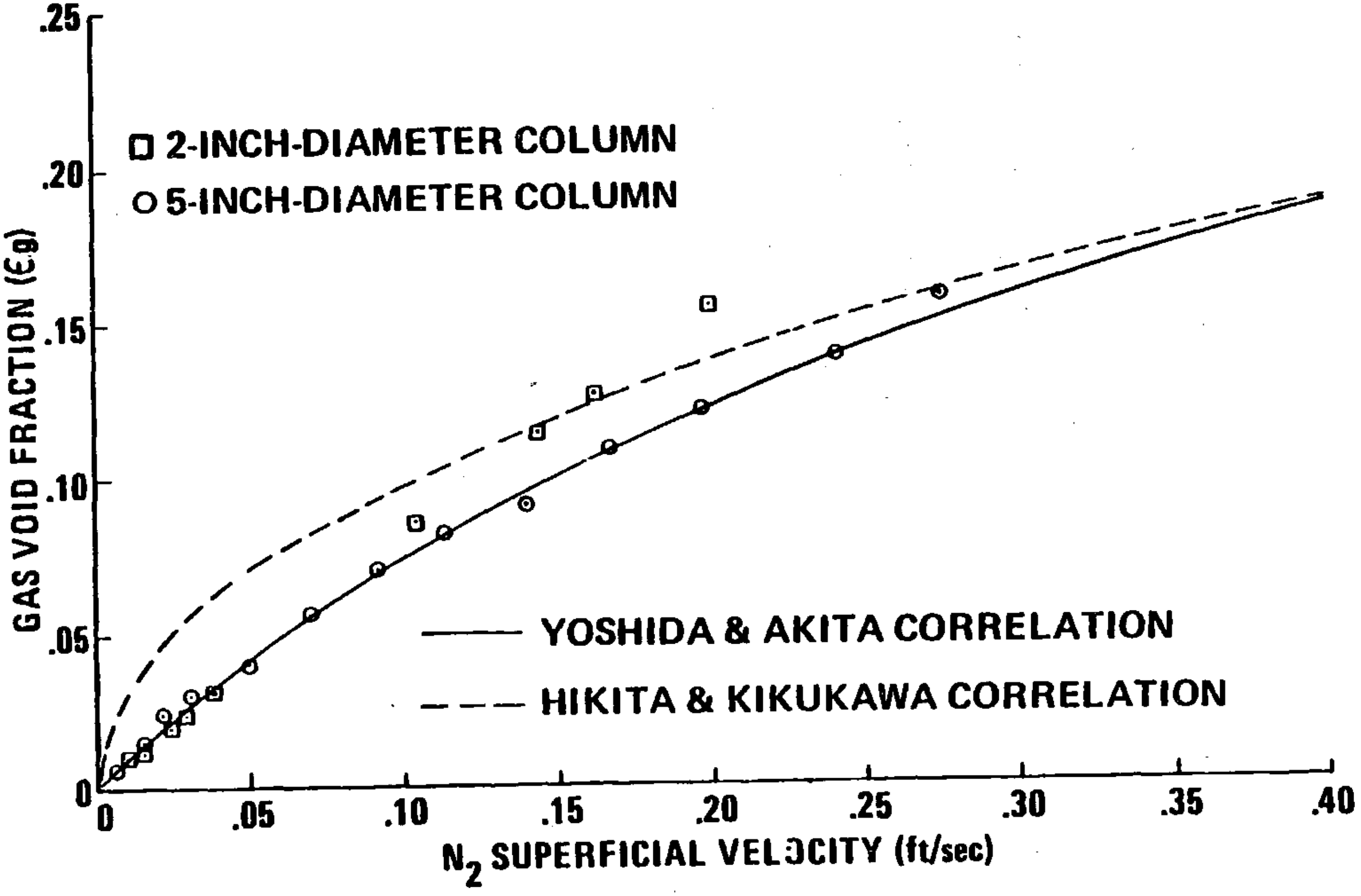


Figure (2)

Gas Holdup in Methanol/Nitrogen System

- 2-inch-diameter column
- 5-inch-diameter column

METHANOL/NITROGEN

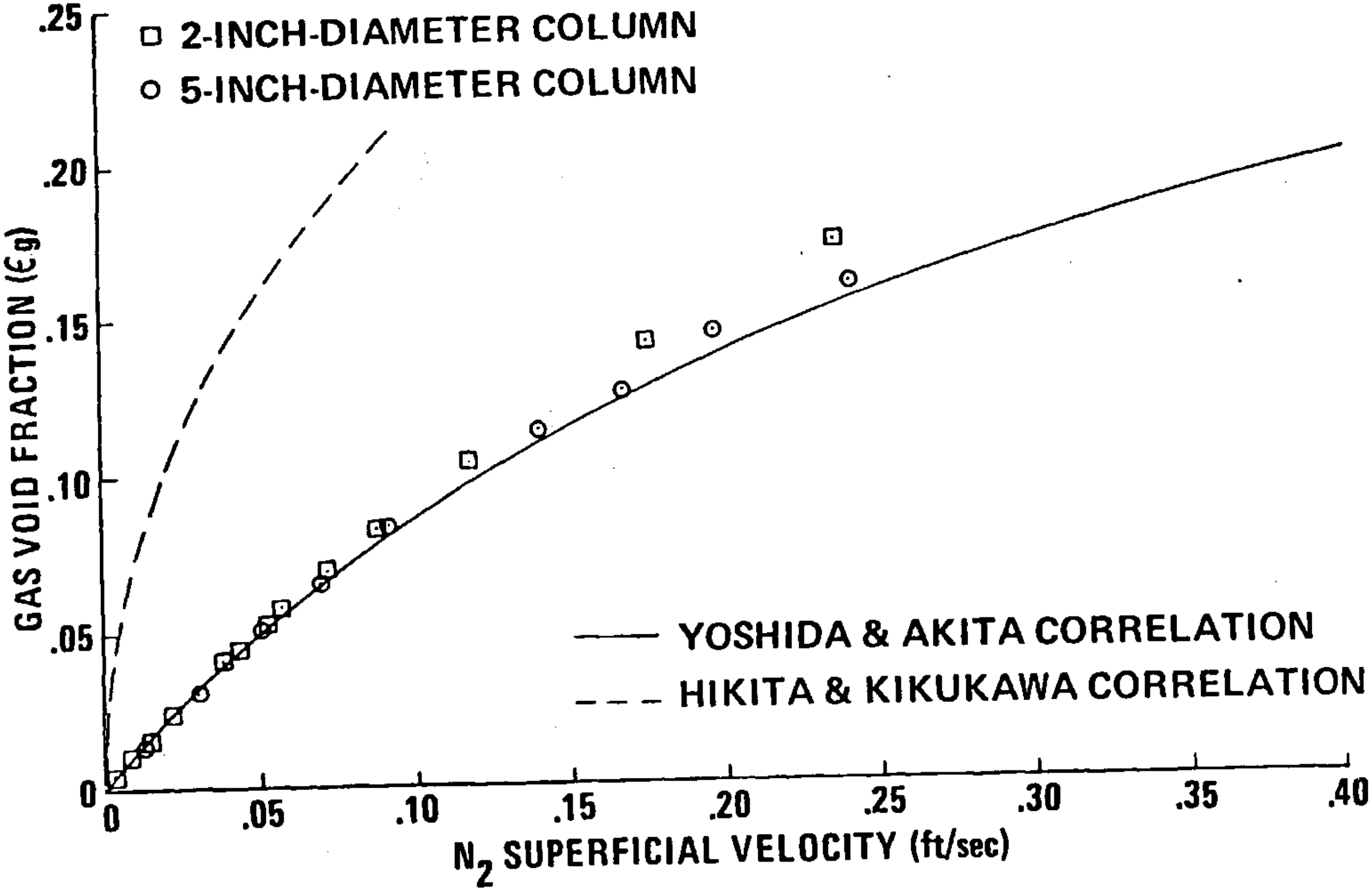


Figure (3)

Effect of Distributor Plate on Gas Holdup
in a 5-inch-diameter Column

5-INCH DIAMETER COLUMN
WATER/NITROGEN

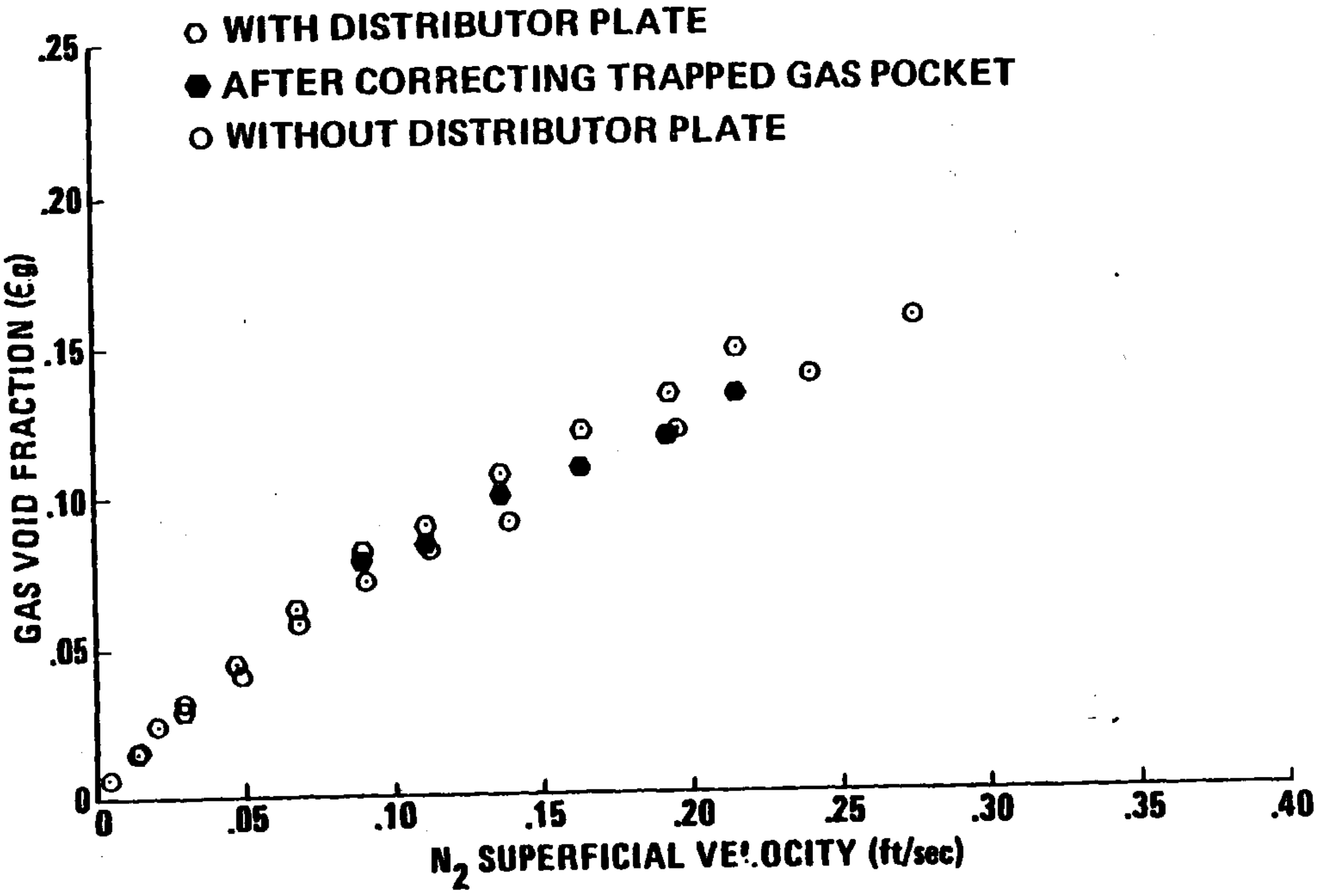


Figure (4)

Effect of Solids on Gas Holdup in a
2-inch-diameter Column

2-INCH-DIAMETER COLUMN

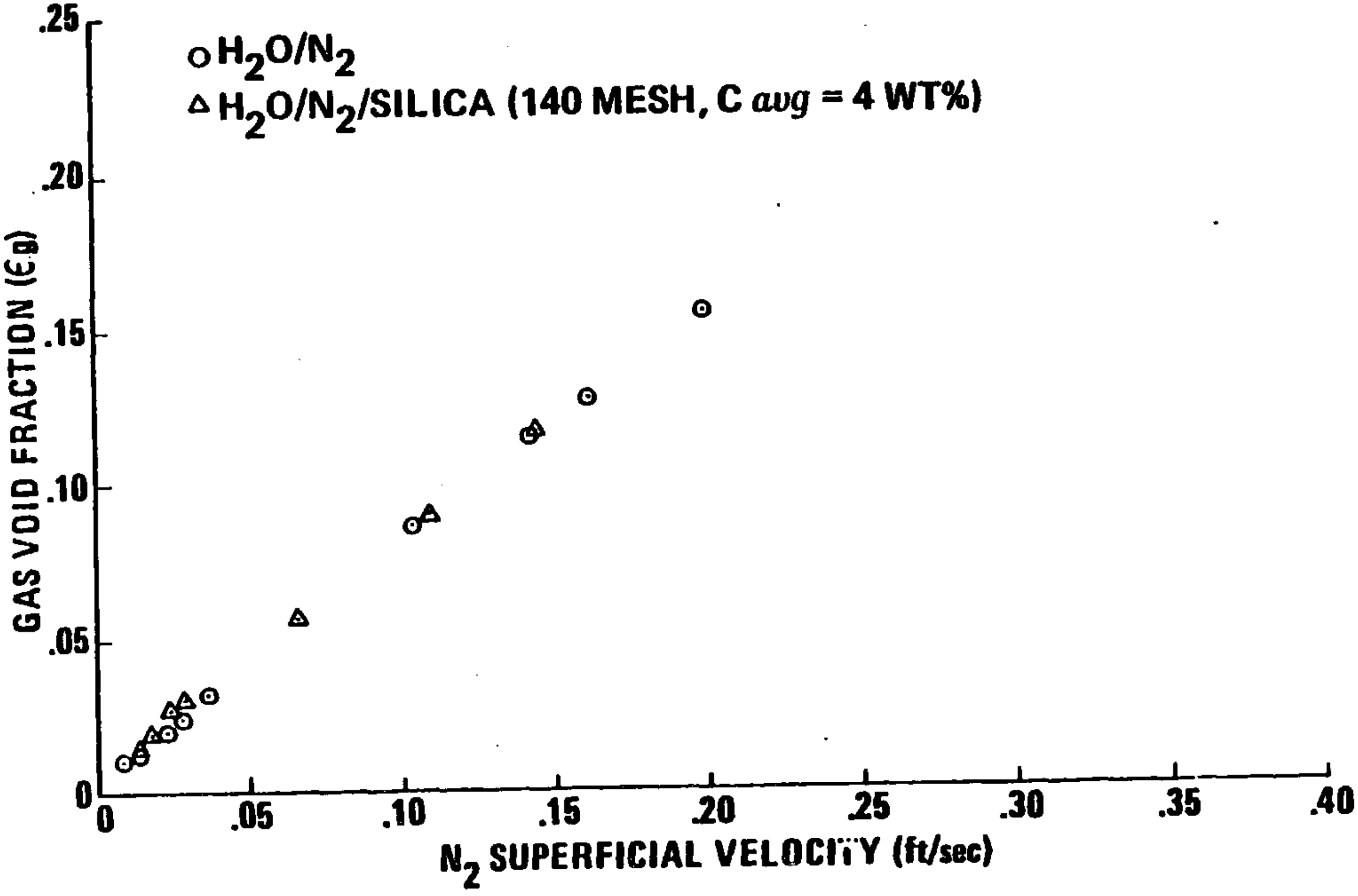


Figure (5)

Effect of Solid Concentration on Gas Holdup
in a 5-inch-diameter Column

5-INCH-DIAMETER COLUMN

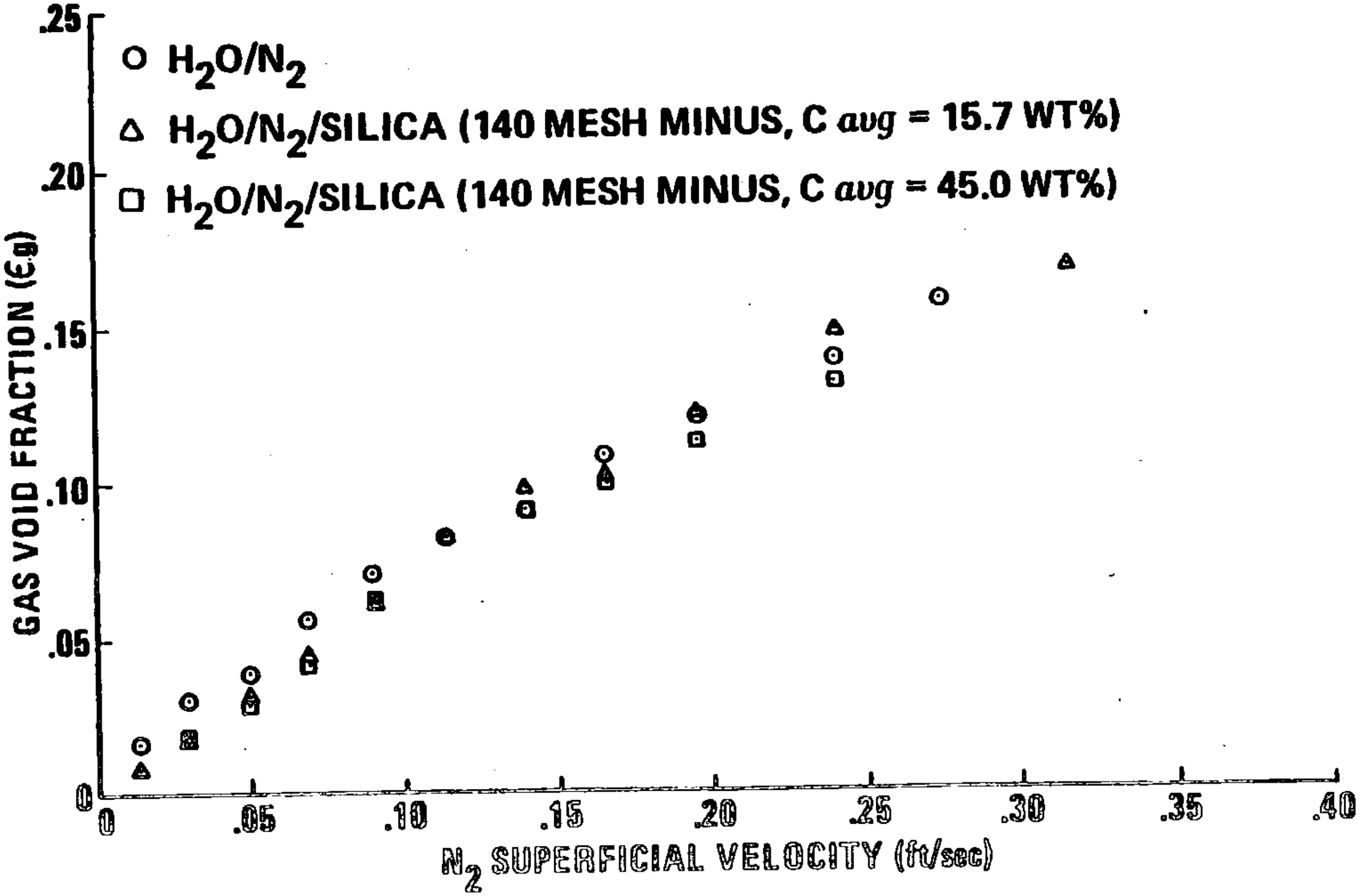
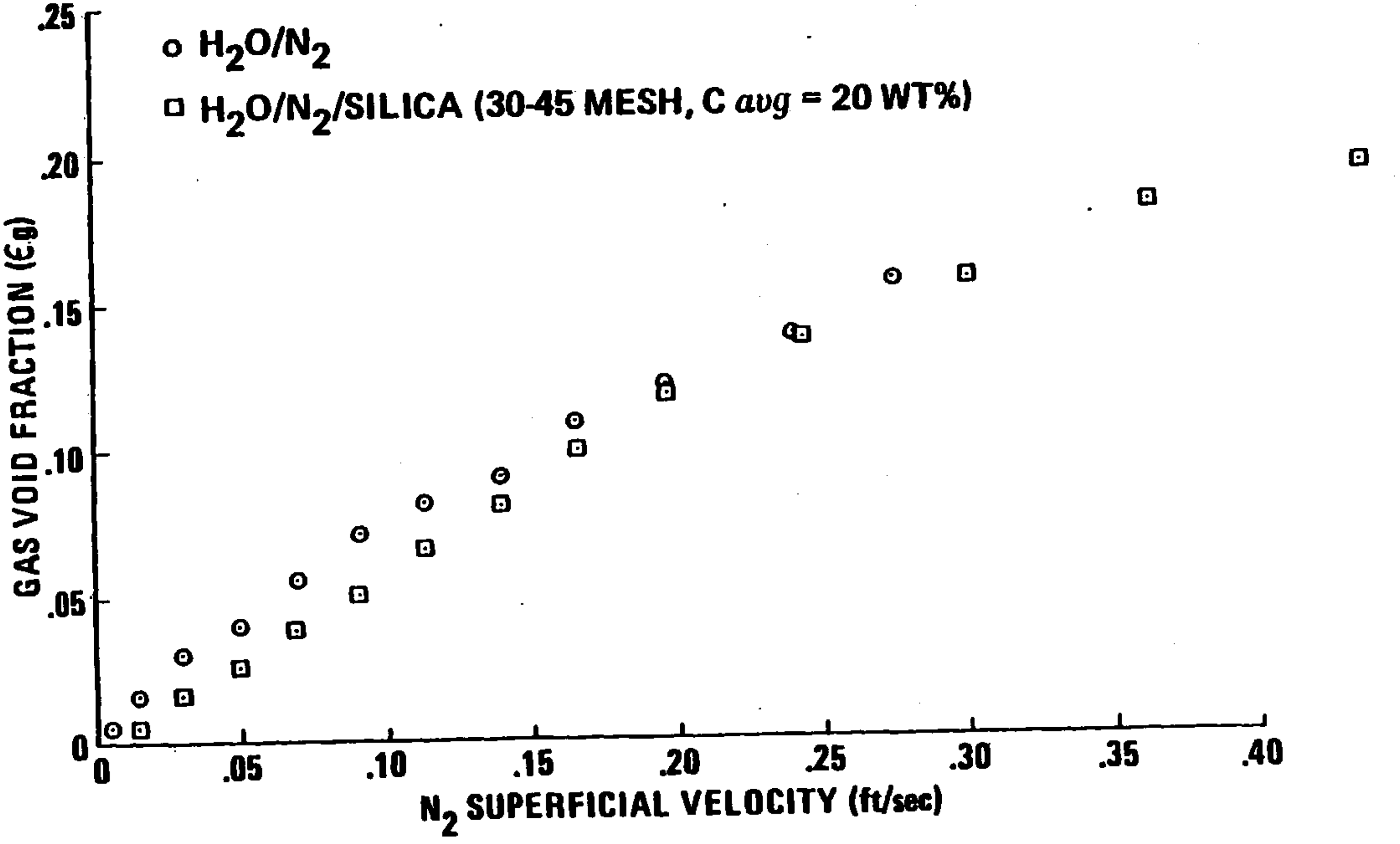


Figure (6)

Effect of Large Solid Particles (30-45 mesh) on Gas Holdup

in a 5-inch-diameter Column

5-INCH-DIAMETER COLUMN



DISSOLVER COLD-FLOW MODELLING

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8. Folder, "Documentation on Project 87-1-X921," contains: Program Authorization, Quarterly Reports, Summaries, and Miscellaneous Memoranda.

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