Part C:

HYDRODYNAMIC BEHAVIOR OF MULTIPHASE REACTORS

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This progress report covers the work done between September 10 , 1981, and September 30, 1981. The report is divided into four sections. The first section deals with a cocurrent bubble column. The second section covers data collected on a batch bubble column. The third section deals with a stirred vessel system, while the fourth deals with a downflow bubble column.

### 1.0 COCURRENT CONTINUOUS BUBBLE COLUMN

As indicated in the last report, the effect of non-Newtonian fluids on the gas holdup and the dispersion coefficient has not been extensively studied yet. Ulbrecht and Baykara (1.1) have recently reported the effect of non-Newtonian medium on the axial dispersion coefficients, but their analysis is restricted to dilute polymer solutions. In the last report, the data collected for CMC (carboxy methyl cellulose) solutions are discussed. This report covers the work done between September 10, 1981 and September 30, 1981. In this period, different PAA (polyacrylamide) solutions have been tested to continue to study the effect of non-Newtonian fluids on the gas holdup and the axial dispersion coefficients. Many investigators (1.2,1.3) have reported the effect of PAA on mass transfer coefficients in a stirred vessel. They have reported that PAA solutions behave differently than CMC solutions because of the viscoelastic behavior present in PAA solutions.

The physical properties of the solutions studied are listed in Table 1.1.

TABLE 1.1
PHYSICAL PROPERTIES OF PAA SOLUTIONS

| Concentration | $\begin{gathered} \rho_{\mathrm{L}} \\ (\mathrm{~g} / \mathrm{cc}) \end{gathered}$ | oL (dynes/cm) | $\begin{array}{r}K \\ c P \\ \hline\end{array}$ | $\underline{1}$ | $\begin{array}{r}\lambda \\ \text { sec } \\ \hline\end{array}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 50 ppm | . 994 | 72.5 | . 97 | 1 | 0 |
| 200 ppm | . 994 | 71.4 | 1.47 | 1 | 0 |
| $500 \mathrm{ppma} 40^{\circ} \mathrm{C}$ | . 994 | 70.6 | 1.84 | . 977 | . 32 |
| ( $25^{\circ} \mathrm{C}$ | . 996 | 70.6 | 2.56 | . 973 | . 32 |
| $1000 \mathrm{ppmo} 40^{\circ} \mathrm{C}$ | . 994 | 71.4 | 3.42 | . 96 | . 5 |
| ( $25^{\circ} \mathrm{C}$ | . 998 | 71.4 | 3.8 | . 948 | . 5 |
| $2000 \mathrm{ppm} @ 40^{\circ} \mathrm{C}$ | . 994 | 69.2 | 4.0 | . 931 | 1.1 |
| @ $25^{\circ} \mathrm{C}$ | . 998 | 69.2 | 4.42 | . 923 | 1.1 |
| 3000 ppm - $40^{\circ} \mathrm{C}$ | . 995 | 70.4 | 6.2 | . 886 | . 89 |
| @ $25^{\circ} \mathrm{C}$ | . 999 | 70.4 | 7.6 | . 845 | . 89 |
| 4000 ppm @ $40^{\circ} \mathrm{C}$ | . 995 | 67.3 | 7.7 | . 93 | . 97 |
| ( $25^{\circ} \mathrm{C}$ | . 999 | 67.3 | 8.8 | . 92 | . 97 |

The viscoelasticity is manifested in the values of relaxation time or $\lambda$. When a viscoelastic liquid flows, only a part of the energy expended will dissipate through viscous friction. The rest will be stored in the liquid and then released as soon as the liquid comes to rest. This process can be characterized with the help of relaxation time. The relaxation time is calculated by the method reported by Yagi and Yoshida. (1.2) The values are approximate and deserve further investigation.

### 1.1 Results and Discussion

The gas holdup values are calculated with the help of the hydrostatic technique as mentioned in the last report. The gas holdup values showed an increase with an increase in the gas velocity. Figure 1.1 shows the effect of gas velocity on gas holdup for the 50 ppm solution. As can be seen, the effect of liquid velocity is negligible. Figure 1.2 shows the same effect for 2000 ppm solution. It can be seen that the effect of liquid velocity is significant. This is believed to probably be a result of the relaxation time. A rising cloud of bubbles dissipates the energy into the surrounding liquid due to viscous friction. The viscoelastic behavior can show a significant effect only if this process takes place over a time scale comparable to the relaxation time. It is believed that the bubble rise velocity in the churn-turbulent regime is in the range of $60-80 \mathrm{~cm} / \mathrm{s}$. Therefore, the average residence time of bubbles would be $4-5 \mathrm{sec}$. Relaxation time for higher ppm solutions is in the range of 1 sec . and can therefore show some effect.

It should be noted here that the viscosity of the solution did not show any significant effect. On the other hand, holdup increased with an increase in PAA concentration. Visual observations revealed that higher PAA concentrations showed foaming characteristics. As mentioned in the earlier report, surface-active agents can play an important role in determining the values of gas holdup without changing the physico-chemical properties of the solution significantly. The foaming characteristics is an indication of the presence of surface-active agents. Probably the surface agents more than offset the effect of the viscosity. Similar to the alcohol solutions, after


GAS HOLDUP VS SUPERFICIAL GAS VELOCITYISO PPM PAA SOLUTIONJ
FIGURE 1.1


FIGURE 1.2
certain additions, the effect diminished and further addition of polymer did not cause any additional increase in the holdup values. The holdup values are compared with the values obtained with the help of different available correlations in the literature and attached in the appendix.

To see the flow regime characteristics of the system, drift flux as a function of gas holdup plots are prepared. One of the graphs is shown in Figure 1.3 which is for 1000 ppm solution. The graph reveals that most of the data lie in the churn turbulent regime. Similar observations are made for other concentrations. It was decided to use Zuber-Findley's(1.4) approach for the evaluation of flow regime characteristics. This equation can be written as

$$
\begin{equation*}
\frac{v_{G}}{E_{G}}=c_{1}+c_{0}\left(v_{G}+v_{L}\right) \tag{1.1}
\end{equation*}
$$

The Zuber-Findley coefficients are listed in Table 1.2.

TABLE 1.2
ZUBER-FINDLEY'S COEFFICIENT FOR PAA SOLUTIONS

| Concentration | $C_{0}$ | $\underline{C}_{1}$ |
| ---: | :--- | :--- |
| 50 ppm | .105 | 2.89 |
| 200 ppm | .11 | 2.55 |
| 500 ppm | .09 | 2.60 |
| 1000 ppm | .11 | 2.43 |
| 2000 ppm | .083 | 2.13 |
| 3000 ppm | .106 | 2.04 |
| 4000 ppm | .121 | 1.94 |

Figure 1.4 shows the graph of $\frac{V_{G}}{E_{A}}$ vs $\left(V_{G}+V_{L}\right)$ for 2000 ppm solution. It can be seen that most of the points lie on a straight line. ZuberFindley's coefficient shows that $C_{0}$ remains fairly constant indicating that bubble size does not change significantly with change in the concentration. The value of $C_{0}$ decreases with an increase in the concentration. This is an indication that an increase in holdup value is a direct result of the uniform


radial distribution, as it is known that $C_{0}$ is a nonuniform distribution parameter. It should be noted that for CMC solutions the $C_{1}$ values showed considerable increase with increase in concentration, indicating an increase in the bubble sizes. Probably, in the case of PAA solutions, the surface active agents kept the bubble size constant.

The axial dispersion coefficients showed very strange behavior. Figure 1.5 shows the dispersion coefficient values for 200 ppm solution as a function of gas velocity. It can be seen that the effect of liquid velocity is negligible. As the concentration increased further, the dispersion coefficients showed decrease in the value for 2000 ppm solution and the dispersion coefficients showed a minimum. This can be explained on the basis of the relaxation time. It is believed that the energy is dissipated in the forin of recoiling of liquid in the case of viscoelastic liquids. This results in less energy dissipation in liquid recirculation which is a main contributing factor for the axial backmixing. Figure 1.6 shows the axial dispersion coefficient values for 4000 ppm solution. It can be seen that the effect of liquid velocity is significant. It reveals that as the liquid velocity is increased, the dispersion coefficient increases. This surprising trend is observed only for 3000 ppm and 4000 ppm solutions. The reasons for this strange behavior cannot be explained yet.

DISPERSION COEFFICIENT VS SUPERFICIAL GAS VELOCITY[200 PPM PAA]
FIGURE 1.5


1.1 Ulbrecht, J. J., and Z. S. Baykara, Paper presented at ACS Meeting, Las Vegas, August, 1980.
1.2 Yagi, H., and F. Yoshida, Ind. Eng. Chem. Process Des. Dev., 14 (4), 488, 1975.
1.3 Ranade, V. R., and J. L. Vibrecht, AlChE J., 24 (5), 796, 1978.
1.4 Zuber, N., and J. A. Findley, J. of Ht. Transfer (Trans. ASME), ser. C, 87, 453, 1965.

APPENDIX 1.1




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| :--- | :--- | :--- |
| $0.0058 \varepsilon$ | 0.24182 | 0.18816 |
| 0.00556 | 0.24311 | 0.18213 |
| 0.00533 | 0.24406 | 0.17229 |
| 0.00516 | 0.24493 | 0.17504 |
| 0.00501 | 0.24547 | 0.16323 |
| 0.00450 | 0.24601 | 0.15947 |
| 0.00484 | 0.24625 | 0.15714 |
| 0.00637 | 0.23997 | 0.21808 |
| 0.00589 | 0.24177 | 0.18982 |
| 0.00558 | 0.24304 | 0.18352 |
| 0.00535 | 0.24398 | 0.17912 |
| 0.00518 | 0.24473 | 0.17180 |
| 0.00504 | 0.24535 | 0.16845 |
| 0.00492 | 0.24589 | 0.16424 |
| 0.00487 | 0.24612 | 0.16067 |

58.25490
$201.34 ¢ 52$
281.46316
332.42544
366.67966
421.52373
458.59135
475.32479
164.08541
157.97220
255.34403
316.54637
360.87636
403.14700
439.10595
454.12368



$$
\begin{aligned}
& 180.44041 \\
& 227.32590 \\
& 269.12038 \\
& 302.90367 \\
& 335.87586 \\
& 362.88122 \\
& 388.76715 \\
& 100.90049 \\
& 177.83392 \\
& 211.84297 \\
& 251.95094 \\
& 290.84553 \\
& 320.58539 \\
& 348.93593 \\
& 374.26786 \\
& 385.45318
\end{aligned}
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### 2.0 BATCH BUBBLE COLUMN

The holdup characteristics of three-phase systems are studied in highly viscous non-Newtonian medium. Kim et al. (1975, 1977) have analyzed phase holdup and bubble characteristics in three-phase fluidized beds with various non-Newtonian (carboxymethylcellulose solutions) and Newtonian solutions. For particles of sizes in the range of $1-6 \mathrm{~mm}$, and gas velocity between $0-0.1 \mathrm{~m} / \mathrm{s}$, Kim et al. (1975) found a slight decrease in $\varepsilon_{\ell}-\varepsilon_{G}$ with respect to gas velocity. Kim et al. (1975) have reported that bubtle sizes are relatively insensitive to viscosity and surface tension for similar systems with a $U_{G}<0.06 \mathrm{~m} / \mathrm{s}$. To study the effect of viscosity on the bubble size distribution in three-phase systems, slurries of sand and polystyrene are studied. Properties of the sand and polystyrene slurries are given in Table 2.1. Phase holdups are measured by a hydrostatic head method, and relative bubble sizes and rise velocities are determined using the dynamic gasdisengagement method.

For the three-phase experiments with water as a liquid medium, a distribution of solids along the column is observed. For, polystyrene runs, gas holdup varied axially with a deviation of less than 3\%. Tables A2.1 to A2.4 indicate the comparison of phase holdups for $10 w t \%$ sand/10, 20 , and 30 wt\% polystyrene in water. For the run of air-water-sand, a significant solid distribution is observed, with part of the sand settling into the conical section of the column. For CMC solution-solid runs, no axial variation of phase holdups is observed. Tables A2.5-A2.8 indicate that correlation of Kito et al. (1976) is applicable to air-water-solid runs. Begovich and Watson's (1978) correlation predicts very low gas holdup for air-water-solid systems; whereas, for air-CMC solution/solid runs it predicts values of gas holdup within a reasonable agreement.

From Figure 2.1, it is clear that the gas holdup is virtually unaffected by the concentration of polystyrene and there is negligible effect of the presence of solids on the gas holdup. It should be noted that the


TABLE 2.1
PHYSICAL PROPERTIES OF THE SLURRIES

| Solid | Wt\% of Solid | $\begin{array}{r} \rho \mathrm{L} \\ \mathrm{gm} / \mathrm{c} \mathrm{C} \\ \hline \end{array}$ | $\begin{gathered} \sigma \\ \text { Dynes/cm } \\ \hline \end{gathered}$ | Consistency Index $C P(\sec )^{n-1}$ | Flow Behavior Index (n) |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Sand* | 10\% | 1.0 | 72.0 | 1.0 | 1.0 |
| Polystyrene** | 10\% | 1.0 | 72.0 | 1.0 | 1.0 |
| Polystyrene | 20\% | 1.0 | 72.0 | 1.0 | 1.0 |
| Polystyrene | 30\% | 1.0 | 72.0 | 1.0 | 1.0 |
| Polystyrene*** | 10\% | 0.9997 | 68.2 | 311 | 0.968 |
| Polystyrene*** | 20\% | 0.9997 | 67.5 | 438 | 0.946 |
| Sand*** | 10\% | 0.9997 | 69.2 | 169 | 0.914 |
| Sand*** | 20\% | 0.9997 | 69.1 | 119 | 0.952 |

* All sand used has an average size (120 $\mu \mathrm{m}$ ) and a density of $2.65 \mathrm{gm} / \mathrm{cc}$. ** All polystyrene used has an average size ( 320 m ) and a density of $1.2 \mathrm{gm} / \mathrm{cc}$.
*** These slurries contain $0.5 \mathrm{wt} \%$ CMC solution as liquid phase.
calculation of gas holdup is based on the assumption of no axial variation of gas holdup. The gas holdup in a highly viscous, non-Newtonian solution has been correlated by

$$
\begin{aligned}
& \qquad \varepsilon_{G}=0.287 V_{G}^{0.536}\left(\mu_{e f f}\right)^{-0.121} \\
& \text { Where, } \mu_{e f f}=k\left(5000.0 \times V_{G}\right)^{n-1}\left(V_{G} \text { in } \mathrm{m} / \mathrm{s}\right) \text {. Figures } 2.2 \text { and } 2.3 \\
& \text { indicate the gas holdup data for } 10 \text { and } 20 \mathrm{wt} \% \text { polystyrene and sand in } C M C \\
& \text { solutions. The air-CMC solution curve is based on the equation reported } \\
& \text { above. For polystyrene-CMC solution mixtures, the gas holdup tends to lie } \\
& \text { above the air-CMC solution line; whereas for sand, it lies below. The effect } \\
& \text { of the addition of solids on gas holdup in } C M C \text { solutions is more than air- }
\end{aligned}
$$ water-solid runs.




Superficial Gas Velocity, m/sec

Figure 2.4 shows gas holdup data obtained from dynamic gas the disengagement method for air-water-polystyrene $\varepsilon_{G}$ as well as $\varepsilon_{G, s}$ (gas holdup due to small bubbles) decreases with an increase in concentration of solids. $\varepsilon_{G, \ell}$ (gas holdup due to large bubbles) shows a pronounced increase with the concentration of solids. Figure 2.5 shows bubble rise velocities as a function of superficial gas velocity. $U_{b r, s}$ (the bubble rise velocity of small bubbles) is virtually independent of gas velocity, although it shows a definite increase with solid concentration.

Figures 2.6 and 2.7 shows $\varepsilon_{G} V_{S} V_{G}$ and $U_{b r}-V_{G}$ for air-CMC solutionpolystyrene system. With the addition of solids, $\varepsilon_{G, s}$ increases; whereas $\varepsilon_{G, \ell}$ shows a significant decrease. This indicates that with the addition of solids, smaller bubbles are formed. On the other hand, in the air-waterpolystyrene system, $\varepsilon_{G, s}$ shows a decrease with $\varepsilon_{G, \ell}$ increasing when polystyrene concentration is increased from $10 \%$ to $30 \%$. Since interfacial area is mostly determined by small bubbles, with the addition of polystyrene "a" should increase in the mixture of air-CMC solution-polystyrene. From Figure 2.7, it can be seen that $U_{b r, s}$ increases with the addition of solids. It seems strange that $U_{b r, s}$ as well as $\varepsilon_{G, s}$ increases with the addition of solids, but it is possible as the bubble size distribution itself is greatly altered. The bubble rise velocities of large bubbles are not shown in the figure, as a small error in the slope of plot of $\varepsilon_{G}$, ( $t$ ) vs time can produce large errors in the predicted value of $U_{b r, \ell}$. This is especially true for three-phase systems or highly viscous solutions. $U_{b r, \ell}$ is generally greater than $1.0 \mathrm{~m} / \mathrm{s}$.

More data are needed before any conclusions can be drawn about bubble size distribution, "a" and $K_{L}$ a in three-phase systems. The dynamic gas disengagement method in conjunction with the knowledge of "a" and $K_{L}$ a can be a great tool in the explanation of observed trends.



FIGURE 2.5



FIGURE 2.7
2.1 Kim, S. D., C. G. J. Baker, and M. A. Bergougnou, "Phase Holdup Characteristics of Three-Phase Fluidized Beds," The Canadian Journal of Chem. Eng., 53, 134 (1975).
2.2 Kim, S. D., C. G. J. Baker, and M. A. Bergougnou, "Bubble Characteristics in Three-Phase Fluidized Beds," Chem. Ena. Sci., 32, 1299 (1977).
2.3 Begovich, J. M., and J. S. Watson, "Hydrodynamic Characteristics of Three-Phase Fluidized Beds," Fluidization, Proceedings of the Second Engineering Conference, Trinity College, Cambridge, England, April 2-6, 1978, edited by Davidson, J. F., and D. L. Keairns, 190-195 (1978).
2.4 Kito, M., T. Shimida, T. Sakai, S. Sugiyama, and C. Y. Wen, Fluidization Technology, edited by Keairns, D. L., Volume 1, Washington Hemisphere Publishing Company (1976).
TABLE A2. 1

TABI F A2.?
EXPERIMENTAL GAS-LIQUTO-ECLIE IIDLDUPS CEMPAFED EITH EXISTIMG CORRELATICRS












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TARLF A2. 3


TABLE A2． 4




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TABLE A2.5


TABLE A2. $\bar{O}$


TABLE A2.7
EXEEHIHENTAL GAS-LIQUIC-SCLID IIILDUFS CCREARED WITH EXISIING CORKELATICNS


## TABLE A2. 8





### 3.0 MECHANICALLY AGITATED VESSEL

### 3.1 Introduction

Liquid phase viscosity is a very important parameter which affects the mass transfer coefficient significantly in gas-liquid and gas-liquid-solid agitated reactors. However, no systematic studies are reported in literature in this area. Only one work reported is by Elstner and Onken, (3.1) who have obtained a mass transfer coefficient, $k_{\ell}$, for glycerin solutions of various concentrations. However, their viscosity range studied is very narrow, and they have not measured or estimated the power consumption per unit liquid volume. Hence, in this work data of mass transfer coefficients and power/ volume in an agitated reactor system for glycerin solutions of various viscosities were obtained.

The advantage of using glycerin solutions is that glycerin is a Newtonian fluid, and it can provide a wide range of liquid phase viscosity, but its surface tension remains almost constant. For these experiments, oxygen was absorbed in glycerin solutions of various concentrations. The viscosity of the solution was measured by Brookfield LVT type viscometer.

### 3.2 Resuits and Discussion

Since the geometry of our system is peculiar and does not conform to the standard configuration (for example, we utilize two impellers having the ratio of impeller diameter-to-vessel diameter of 0.57 ), it was decided to initially obtain some data in a standard vessel arrangement (only one impeller, ratio of liquid height-to-vessel diameter equal to 1 , and impeller diameter-to-vessel diameter $=0.45$ ) so as to give a good comparison of our data with that of Elstner and Onken. (3.1) Data for $10 \mathrm{wt} \%$ and $40 \mathrm{wt} \%$ glycerin solutions were obtained in standard vessel configuration and are reported in Table 3.1. They are plotted in Figure 3.1. In these measurements, $k_{\ell}{ }^{a}$ was first evaluated. To evaluate "a", the surface area was taken to be the crosssection of the vessel. Knowing $k_{\ell} a$ and " $a$ ", $k_{\ell}$ was evaluated. The figure

## TABLE 3.1

## MASS TRANSFER COEFFICIENT $k_{\ell}$ FOR STANDARD VESSEL ARRANGEMENT

Temperature - $24^{\circ} \mathrm{C}$ (average)

| rpm | $k_{\ell} \times 10^{-3}(\mathrm{~cm} / \mathrm{sec})$ |  |  |
| :---: | :---: | :---: | :---: |
|  | Water | $10 \mathrm{wt} \%$ Glycerin | 40 wt\% Glycerin |
| 400 | 5.3 | 2.7 | 1.4 |
| 600 | 8.5 | 4.0 | 1.7 |
| 800 | 11.5 | 6.8 | 2.3 |

also shows the data of Elstner and Onken under similar conditions. Even though our data values are slightly higher than those of Elstner and Onken, considering the fact that the two methods of measurement are entirely different (we employ measurement of change in total pressure of the gas phase with respect to time for a batch system, whereas Elstner and Onken employ an oxygen concentration measurement cell), the difference in values of $k_{\ell}$ is within acceptable limits. To compare the order of magnitude of values, data of $k_{\ell}$ obtained for water in our system for a standard vessel arrangement are also shown in Figure 3.1.

Table 3.2 summarizes the data of $k_{\ell} a$ for various concentrations of glycerin solutions at three values of rpm. Figures 3.2, 3.3, and 3.4 show the nature of these data on log-log plots. It can be seen that the liquid phase viscosity has a very significant effect on $k_{l} a$. With an increase in viscosity, the $k_{\ell} a$ value decreases several-fold for all three values of rpm. In Figure 3.4, data for the oxygen-water system are also shown to give a comparison about the order of magnitude of $k_{\ell}$ a values.

The data of power consumed per unit liquid volume for the above experiments are summarized in Table 3.3 and are plotted in Figures 3.5 and 3.6. From Figure 3.5, the slopes of the lines for various values of viscosity indicate that for higher values of viscosity, increase in power input increases the value of $k_{\ell^{a}}$ significantly as compared to lower values of viscosity. Figure 3.6 indicates that as compared to higher values of rpm, for lower values of rpm, the increase in power consumed is much higher with increase in viscosity.


FIGURE 3.1 Values of Mass Transfer Coefficient ' $k_{l}$ ' for Various Values of Agitator RPM in Standard Vessel Arranqement

TABLE 3.2

## VALUES OF $k_{\ell}$ a FOR GLYCERIN SOLUTIONS <br> Temperature - $24^{\circ} \mathrm{C}$ (average)

| Glycerin Concentration | Viscosity | $k_{\text {g }}{ }^{\text {a }}\left(\mathrm{sec}^{-1}\right)$ |  |  |
| :---: | :---: | :---: | :---: | :---: |
| Volume \% | cP | 1000 rpm | 800 rpm | 600 rpm |
| 51 | 7.0 | 0.08627 | 0.0296 | 0.0042 |
| 58 | 10.6 | 0.04624 | 0.0103 | 0.0020 |
| 70 | 20.0 | 0.02598 | 0.00675 | 0.000832 |
| 86 | 66.0 | 0.01835 | 0.0051 | 0.00050 |
| 92 | 92.0 | 0.0087 | 0.0030 | 0.00040 |
| 94 | 115.0 | 0.00845 | 0.0017 | 0.00032 |



FIGURE $3.2 \quad$ ' $k_{e} a^{\prime}$ as a Function of Liquid Phase Viscosity



FIGURE 3.3 ' $k_{\ell} a^{\prime}$ as a Function of Glycerin Concentration


FIGURE 3.4 Variation of ' $k_{\ell}$ a' with Agitator RPM

## Table 3.3

## POWER CONSUMPTION FOR GLYCERIN SOLUTIONS

| Glycerin Concentration | Viscosity | Power/Volume ( $\mathrm{W} / \mathrm{m}^{3}$ ) |  |  |
| :---: | :---: | :---: | :---: | :---: |
| Volume \% | cP | 1000 rpm | 800 rpm | 600 rpm |
| 51 | 7.0 | 6.8 | 3.6 | 1.1 |
| 70 | 20.0 | 8.6 | 5.0 | 1.8 |
| 86 | 66.0 | 9.6 | 6.4 | 2.7 |
| 92 | 92.0 | 11.4 | 7.3 | 3.2 |
| 94 | 115.0 | 13.6 | 8.2 | 4.1 |

## LEGEND



FIGURE 3.5
Variation of ' $k_{\hat{l}}{ }^{\text {a' }}$ with Power/Volume


FIGURE 3.6 Variation of Power/Volume with Viscosity

## References

3.1 Elstner, F., and U. Onken, Ger. Chem. Eng., 4, 84-89 (1981).

### 4.0 CONTINUOUS COCURRENT DOWNFLOW BUBBLE COLUMN

### 4.1 Introduction

The use of bubble columns has been widely employed in gas-liquid systems and recently to gas-liquid-solid systems in Fischer-Tropsch synthesis, oxidation of organic compounds, and in coal liquefaction. The reported work is exclusively concentration on systems in which gas is introduced at the bottom of the column and liquid may be either in batch mode or flows cocurrently upward along with the gas phase. However, the gas phase residence time is limited due to the rise in velocity of the bubbles, which can be overcome provided the gas is dispersed from the top of the column in a liquid flowing vertically downward, so that the gas bubbles are forced down by the liquid flow in a direction opposite to that imposed by the bouyancy. Under these conditions, the mean residence time of the gas phase can be extended to the point of a state of suspension by variation of the liquid velocity.

The above premise has been substantiated by experimental measurements of phase holdup in a glass column 0.075 m ID and 2.45 m in height. The gas phase holdup was measured using the hydrostatic head technique. The effect of a wide range of physical parameters, such as superficial gas velocity, superficial liquid velocity, surface tension, and electrolyte concentrations, were studied.

The experimental data reported is the progress made during the period mentioned. The results obtained can be explained qualitatively, and a detailed analysis of this data will be provided at a later stage.

### 4.2 Experimental Setup and Procedure

### 4.2.1 Experimental Setup

The downflow bubble column consisted of a glass column with an internal diameter of 0.075 m and height of 2.45 m . The gas phase is introduced through the top of the column. The downflowing gas-liquid or gas-
liquid-solid mixture is discharged into a cylindrical disengaging tank mate of plexiglass with an internal diameter of 0.30 m and height of 0.30 m . The bottom of the disengaging tank is fitted with a conical stainless steel flat circular plate, 0.075 m in diameter, which acts as a baffle. The baffle is located 0.10 m from the bottom section of the cone which prevents any containment of the gas phase in the recycle liquid. The degassed liquid is recycled by means of a slurry pump having a capacity of 40 gpm , while the gas phase is drawn off at the top of the disengaging tank. Two glass bulbs, 0.152 m in diameter, are mounted at the top of the column and the liquid line and serve to disengage any gas which may be entrained in the recycle liquid.

The gas phase used is always air. The air inlet pressure is maintained constant with the help of a pressure regulator. The gas flow rate is monitored with the help of two rotometers of different ranges mounted in parallel. The liquid flow rate is metered using a calibrated elbow meter inthe liquid line which is connected to a liquid indicator. The slurries to be used were metered by using an ultrasonic measuring device.

The column is fitted with six ports along the length; the distance between two consecutive ports is 0.035 m . Four of these ports are used as pressure taps to measure the pressure along the length of the column. The distance between the two pressure taps is 0.61 m . The pressure taps are connected to a mercury manometer, one end open to atmosphere. A back-flushing system is incorporated to ensure that no air bubbles are entrained in the lines connecting the ports to the liquid level indicator. The two other ports were provided for conductivity probes. These probes were at a distance of 1.22 m apart. The conductivity of a two-phase or three-phase mixture depends on the relative amount of each phase present in the mixture. This principle was employed to measure the gas holdup with these probes. The method has been used previously by Stepanek et al. (4.1) However, during the course of the experiments it was found that the gas phase would accummulate in the region directly below the probe at higher gas velocities since the probes resulted in the formation of a "wake" region. The entrapped gas bubbles would coales'. and eventually very large bubbles or slugs would discharge, rising through the column. Hence, the probes have been eliminated and the ports closed.

The system is devoid of any problems such as leaks which prevailed previously in the disengaging section or entrainment of large amounts of the gas phase in the recycle liquid. Figure 4.1 is the process diagram of the cocurrent downflow bubble column. All the experiments were carried out at near-atmospheric conditions and under steady-state conditions.

The surface tension is measured with a Fisher Surface Tensiomat (Model 21) using the du-Nouy method. (4.21) In this method, a platinum ring of precisely known dimensions is suspended from a counterbalanced lever arm. The arm is held horizontal by torsion applied to a taut stainless steel wire to which it is clamped. Increasing the torsion in the wire raises the arm and the ring which carries with it the film of liquid in which it is immersed. The force necessary to pull the test ring free from the surface film is measured directly in dynes/cm. This apparent reading is converted to the absolute value by use of a correction chart.

The viscosity is measured using a Brookfield LVT type viscometer. The spindle is rotated in a given fluid at a constant speed. The torque necessary to overcome the resultant viscous drag is measured. For a given spindle and speed, it produces dial readings proportional to the viscosity. (4.2,4.3)

### 4.2.2 Procedure and Measurement of Holdup

In the case of bubble columns with negligible liquid, the variation of pressure with height is entirely due to the hydrostatic head in gas liquid systems

$$
\begin{align*}
& \frac{d p}{d x}=\varepsilon_{L} \rho_{L}+\varepsilon_{g} \ell_{g}  \tag{4.1}\\
& \varepsilon_{L}+\varepsilon_{g}=1.0 \tag{4.2}
\end{align*}
$$

The above two equations are employed to obtain the gas phase holdup. To use these equations, the manometer readings were first corrected to absolute pressure. However, in the presence of liquid flow, there are two additional terms on the right hand side of Equation (4.1) to account for wall friction and acceleration due to voidage changes along the length of the column. Hence a momentum balance on a control volume changes Equation (4.1) to

$$
\begin{equation*}
\frac{d p}{d x}=\left(\varepsilon_{g} \ell_{g}+\varepsilon_{L} \ell_{L}\right)+q_{L} V_{L}^{2} \frac{d}{d x}\left(\frac{l}{1-\varepsilon_{g}}\right)+\frac{4 T w}{d c} \tag{4.3}
\end{equation*}
$$

The acceleration term is normally small and is neglected; however, in wider columns it can become appreciable. At high flow rates the viscous drag term can account for about as high as $25 \%$ of the first two terms. Hence, the effect of these two terms was first determined. The hydrostatic head was measured in the absence of gas flow through the column for the entire range of liquid velocities covered in this work. It was found that the manometric readings of the liquid hydrostatic head between any two tappings on the column was in very close agreement (within $\pm 2 \%$ ) to the height of liquid between these ports. Also, the axial variation of holdup was found to be negligible since the holdup calculated in the following manner

$$
\begin{equation*}
\varepsilon_{G}=\frac{H H / V G=0^{-H H}}{H H / V G=0} \tag{4.4}
\end{equation*}
$$

between two consecutive tappings was found to be in close agreement along the length of the column. Hence, Equations (4.1) and (4.2) were used to obtain integral values of holdup.

To determine the solid and liquid holdups in three-phase systems, solid-liquid samples will be selected at the differing tappings along the length of the column. By measuring the weight and volume of the slurry, density will be obtained. After filtering and drying the samples, it is possible to calculate the relative volume fraction of liquid and the solid. Using this information, the following time equations will be solved simultaneously to get the values of individual phase holdup as

$$
\begin{align*}
& \varepsilon_{G}+\varepsilon_{L}+\varepsilon_{S}=1.0  \tag{4.5}\\
& \frac{d p}{d x}=\varepsilon_{G} \rho_{G}+\varepsilon_{L} \rho_{L}+\varepsilon_{S} \rho_{S}  \tag{4.6}\\
& \varepsilon_{S} / \varepsilon_{L}=\text { Known quantity } \tag{4.7}
\end{align*}
$$

### 4.3 Results and Discussion

### 4.3.1 Gas Holdup

Gas holdup shows an increase with an increase in the gas velocity but shows a decrease with an increase in liquid velocity (as can be seen clearly in figure 4.2 for air-water data). The experiments were carried out at near-atmospheric pressure at gas velocities ranging from $0.06-2.2 \mathrm{~cm} / \mathrm{s}$ and liquid velocities ranging from $20.0-32.0 \mathrm{~cm} / \mathrm{s}$. The range of gas velocities is extremely low; however, the gas holdup is nearly an order of magnitude greater than in conventional bubble columns operated cocurrently. $\mathrm{Hills}(4.5)$ has reported holdup measurements in a bubble column at high liquid throughputs. At the highest gas velocity, his correlation gives values of gas holdup in a cocurrently operated upward bubble column of less than $1 \%$ for all the liquid velocities employed in this work. The termination points on the curves in the direction of increasing gas holdup represents the limits of the mode of downflow operation within $10 \%$ of the maximum gas velocity which can be employed. This results from the formation of bubble agglomeration at the top of the column due to the migration of large bubbles or slugs formed due to coalescence at the bottom of the column.

### 4.3.2 Effects of Surface Tension and Alcohol Property

The gas holdup decreases with the addition of surfactants such as alcohols $\left(C_{1}-C_{4}\right)$ as compared to the gas holdup obtained in an air-water system as can be seen from Figures 4.3-4.8. However, the effect of alcohol concentration (or surface tension) on the gas holdup is observed to be insignificant (Figures 4.9-4.11); but the effect of the type of alcohol is predominant


FIGURE 4.2


FIGURE 4.3
[\%] dnOרOH S*9


FIGURE 4.5
[Z] dnOרOH SVロ


Figure 4.6

FIGURE 4.7
[\%] dnaרOH S*9



FIGURE 4.9
[\%] dnaาOH SV5

[\%] dnaTOH SVפ


FIGURE 4.11
especially at low liquid velocities (Figure 4.9) as compared to the higher liquid velocities (Figures 4.10 and 4.11). The gas holdup decreased in the following order:

## Methanol < Ethanol < Propanol < Butanol

The effect of surfactants is still ambiguous. The reported literature is at times in complete agreement. Bolton et al. (4.6) and Miller (4.7) reported no effect of surface tension; Schugerl et al., Todt et al., (4.9) and recently Dels(4.8) observed a significant increase in gas holdup with decrease in the surface tension; Sharma et al. (4.10) found only a slight increase in the gas holdup with a decrease in surface tension. Bach and Pilhofer(4.11) made a detailed study on gas holdup characteristics using pure liquids and liquids mixtures. They found that pure liquids and mixtures behaved differently and also that surface tension was found to have no effect on gas holdup in the case of pure liquids. However, Schugerl et al. (4.8) do report a variation in the coalescence behavior of the gas phase in the presence of surfactants. Friedel et al. (4.12) also report a decrease in bubble size from changing from distilled water to tap water to aqueous ethanol. The same behavior was observed during experiments with air-water and air-alcohol systems. However, with different concentrations of butanol, or for different alcohols, significant bubble size variation could not be observed visually.

In alcohol solutions, the coalescence rate is reduced and the bubble size decreases. The decrease in the bubble size results in a decrease in the bouyancy force and a corresponding decrease in the bubble rise velocity. The bouyancy force and the drag force being in opposite directions, for the same liquid velocity, there is a decrease in the gas holdup as bubbles are entrained from the column. A comparison of the data for alcohol solutions and air-water systems reveals this conclusion as can be seen from Figures 4.2 and Figure 4.12. This phenomena is directly opposite to the case of cocurrentlyoperated upflow systems, where the presence of surfactants does increase the gas holdup since the bubble-rise velocity decreases.

In the presence of surfactants, the rise velocity of the same diameter bubble need not be the same. The interface of the bubble is mobile and an internal circulation movement exists in the bubble which reduces the drag on the bubble. 0els et al.(4.13) report that the surfactants are absorbed at the top of the bubble and are transported to the rear, and a surface tension gradient is formed. The surface tension gradient across the bubble depends essentially on the type of alcohol. As the chain length of the alcohol increases, the rigidity of the bubble increases causing a reduction in the bubble rise velocity and hence an increase in gas holdup. For a downflow system, an increase in the chain length of the alcohol should decrease the gas holdup. This behavior is clearly seen in Figure 4.12 at low liquid velocities. However, at higher liquid velocity, the holdup becomes progressively independent of the type of alcohol; since the relative velocity between the two phases is dominated by the liquid velocity (Figures 4.13 and 4.14).

Visual observation indicates the bubble size to be uniform indicating that the flow regime encountered in the range of gas and liquid velocities studied is essentially the bubbly-flow regime. The drift flux diagram in Figures 4.22 and 4.23 clearly shows the absence of any transition from this regime.

The physical properties of the solutions are reported in Table 4.1.

### 4.3.3 Effect of Electrolytes Solution

The effect of electrolyte solutions on the gas holdup has been studied using NaCl in the range of concentrations of 0.05 m to 1.25 m . The gas holdup decreases with an increase in ionic strength up to 0.5 m and is independent of the ionic strength beyond 0.5 m . In upflow systems, Akita and Yoshida(4.14) have reported an overall increase of about $25 \%$ in the holdup on addition of an electrolyte. This increase in voidage is primarily due to tioe postponement of the appearance of large bubbles, since the addition of an electrolyte induces a non-coalescing behavior due to the presence of an icnic. double-polar layer between the gas and liquid phases. Braulich et al. (4.: : report that the holdup is a function of both the concentration and the gas

[\%] dNOרOH SVפ


[¿] dna70H S*5

FIGURE 4.15
[\%] dnaרOH S甘פ

[ $\%$ d dnaloh SV9

FIGURE 4.16


[\%] dnaาOH S甘O
[\%] dnaาOH S*9

[\%] dnaloh S甘פ

## TABLE 4.1

PHYSICAL PROPERTIES: ALCOHOL SOLUTIONS

|  | of $\left(\mathrm{g} / \mathrm{cm}^{3}\right)$ | $\underline{\left.g_{\text {d }} \text { (dynes } / \mathrm{cm}\right)}$ | H ( $C$ ( ${ }^{\text {( }}$ |
| :---: | :---: | :---: | :---: |
| 0.5\% Methanol | 0.994 | 67.96 | 0.83 |
| 0.5\% Ethanol | 0.9931 | 66.96 | 0.83 |
| 0.5\% n-Propanol | 0.9008 | 64.85 | 0.85 |
| 0.5\% n-Butanol | 0.9932 | 60.18 | 0.84 |
| 1.5\% n-Butanol | 0.9912 | 49.316 | 0.85 |
| 3.0\% n-Butanol | 0.9900 | 40.26 | 0.85 |




FIgURE 4.22

velocity. In the present study the holdup decreases with the addition of an electrolyte; however, the decrease in holdup is relatively small. Table 4.2 shows the concentration ranges of NaCl solutions employed and the physical properties of the system.

A visual observation of the systems shows a very uniform bubble size distribution indicating that the bubbly-flow regime is encountered.

Figures 4.15 to 4.20 show the effect of electrolyte concentration.
The experimental data is compared with the correlation for gas holdup provided by Freidal et al.(4.12) Their correlation predicts the holdup to be much higher than the experimental values obtained for the air-water system in our work. The predicted and experimental values are in error by 15-40\% for air-water data. For alcohols and electrolytes, the disagreement is much too large and use of this correlation seems inappropriate. Friedel et al. (4.12) report that their correlation predicts their own experimental data within $28 \%$ standard deviation. The results are presented in Appendix 1. This disagreement may be due to the diameter of the column and nature of the sparger(?) used especially for the air-water system. The holdup is known to be a function of the column diameter up to 0.015 m ID (Akita and Yoshida). (4.14) There is consistent diameter effect where the values of gas holdup in our work $\left(D_{c}=0.075 \mathrm{~m}\right)$, Freidel et al. $(4.12)\left(D_{c}=0.015 \mathrm{~m}\right)$ and Fujie et al. ${ }^{(4.16)}$ ( $D_{C}=0.45 \mathrm{~m}$ ) are compared. The holdup decreases with increase in column diameter in the range of gas and liquid velocities studied.

### 4.3.4 Effect of Viscosity

CMC solutions of 50 ppm and 1000 ppm were employed to determine the effect of viscosity. However, at gas velocities as $10 w$ as $0.06 \mathrm{~cm} / \mathrm{s}$, large gas bubbles and slugs were observed, resulting in a gas cushion at the top of the downflow bubble column at the highest liquid velocity. ( $31.625 \mathrm{~cm} / \mathrm{s}$ ) employed. The appearance of the gas cushion is a sensitive indicator of the heterogeneous flow conditions which prevail and limit the mode of operation of the downflow system. The range of gas velocities were so small that no

TABLE 4.2
PHYSICAL PROPERTIES: NaCl SOLUTIONS

|  | O (dynes/cm) | H (CD) | $\underline{p} \underbrace{\left(g / c m^{3}\right)}$ |
| :---: | :---: | :---: | :---: |
| 0.05 m | 70.50 | 1.0 | 0.998 |
| 0.5 m | 70.15 | 1.22 | 1.0415 |
| 1.0 m | 73.50 | 1.23 | 1.065 |
| 1.25 m | 74.25 | 1.29 | 1.074 |

conclusive data could be obtained, although Freidel et al. (4.12) do rio. measurements of gas holdup in which they varied the liquid velocity $f$ orn 1-11.4 cp. They report a decrease in holdup with an increase in viscosit....
4.4 Proposed Future Work

1. Measurements of gas holdup using solids,
2. Measurement of interfacial areas in gas-liquid and gas-liquia-jolid systems,
3. Analysis and correlation for the data obtained in holdup measurements.

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