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Hydrodynamic study in a slurry-bubble-column reactor

Y. Soong *, F.W. Harke, I.K. Gamwo, R.R. Schehl, M.F. Zarochak

US Department of Energy, Pittsburgh Energy Technology Center, P.O. Box 10940, Pittsburgh, PA 15236, USA

Abstract

Local gas holdup, bubble diameter and bubble rise velocity in the nitrogen/Drakeol-10 oil system were measured at both laboratory (ambient temperature and pressure) and industrially relevant (high temperature and pressure) conditions using a dual conductivity probe in a slurry-bubble-column reactor. It was found that at a constant superficial velocity, the Sauter mean bubble diameter decreases with increasing pressure and temperature. The bubble rise velocity significantly decreases as the pressure increases. Large bubbles rise faster than smaller bubbles. Akita and Yoshida's correlation [1] was utilized to compute the bubble size. Predicted values agree with the experimental data at high temperature.

Keywords: Hydrodynamics; Bubble column reactor

1. Introduction

The Slurry-Bubble-Column Reactor (SBCR) system has been shown to have new applications in the chemical industries and coal liquefaction processing. Slurry-bubble-column Fischer–Tropsch (F–T) reactors have many advantages over other types of F–T reactors. Among them are low capital/operating costs, good temperature control of the F–T reaction, the ability to use a low H_2/CO ratio synthesis gas, low carbon deposition on the catalyst, and small mass-transfer resistances.

To design and efficiently operate a three-phase slurry reactor, it is necessary to understand the hydrodynamic parameters (i.e., bubble size, gas holdup and its axial dispersion, the liquid-side mass transfer coefficient, and the overall heat-transfer coefficient) and the degree of dispersion of the catalyst in the reactor. Both factors within the reactor greatly affect its operation. In general, the presence of small solid particles reduces the gas holdup, due to an increase in the slurry viscosity. The important hydrodynamic factors in the operation of a bubble column reactor are the bubble size, gas holdup, and flow regimes. Extensive work on the hydrodynamic measurements in two-phase systems has been reported in the literature and has been reviewed by several authors [1,2]; however, most of theses studies have been limited to the air–water system. Hydrodynamic measurements using molten wax (typical slurry medium) as liquid medium are rather limited [3–6]. Different hydrodynamic behaviors (e.g., Sauter mean bubble diameter, gas holdup) were reported by these studies for similar physical properties of wax. As indicated by Bukur et al. [3], different waxes, despite similar physical properties, have dissimilar hydrodynamic be-

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[*] Corresponding author. Tel.: (412) 892-4925. Fax: (412) 892-4152.

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haviors. A possible cause for different hydrodynamic behaviors is the flow regime which exists in a particular bubble column. As indicated by Smith et al. [7] at least four flow regimes have been identified: homogeneous bubbling, transition, churn-turbulent, and slug flow. A comprehensive flow regime map for a range of F–T slurry reactor conditions remains to be developed. It has become clear that additional experimental data is required on the hydrodynamic behavior of slurry bubble column reactor systems that employ operating conditions and liquid phase media like those in F–T slurry reactor. This information is necessary for reliable design/scale up of F–T slurry reactors.

2. Experimental

A schematic diagram of the SBCR is shown in Fig. 1. The unit consists of two stainless steel columns, each has an internal diameter of 10 cm and a height of 244 cm. The wax melt column is utilized to store and melt the wax. The main column in which the hydrodynamic studies were conducted has 12 different axial locations for data collection. Experiments were conducted in batch-mode operation (stationary liquid and continuous flow of gas). The feed nitrogen was metered with a mass flow meter, passed through a preheater to the gas distributor, and into the main column. The gas distributor was a perforated plate with 5 × 1-mm holes. The effluent from the reactor was passed through a cold trap, a back-pressure regulator, a wet-test meter, and vented into a hood.

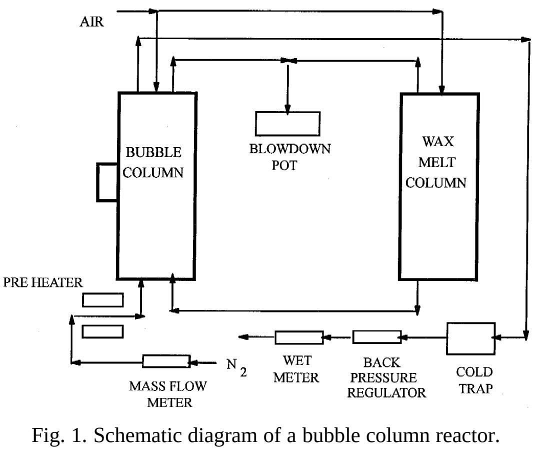

Fig. 1. Schematic diagram of a bubble column reactor.

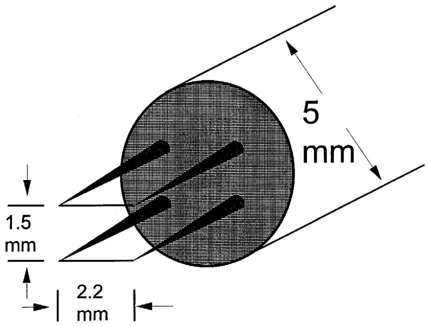

Fig. 2. Schematic diagram of the dual hot-wire probe.

The local gas holdup, bubble diameter, rise velocity, and size distribution were measured by inserting a dual conductivity (hot-wire) probe horizontally into the column at any of the 12 positions and moving it to the desired radial position. A schematic diagram of the dual hot wire probe is presented in Fig. 2. Detailed descriptions of the probe were reported elsewhere [8,9]. The local gas holdup was determined from the ratio of the measured signal's time in the gas phase to its total sampling time at the given location. The bubble diameter was calculated from the time lag in the signals from the two sensing tips (the time difference between the bubble encountering the first hot-wire and the second hot-wire), using the known time of duration of the signals and the distance between the sensing tips [8,9].

The hydrodynamic parameters were measured with the dual hot-wire probe at 30, 50, 71, 111, and 132 cm above the gas distributor in the bubble column. The experiments were conducted at temperatures of 20°C and 265°C, at pressures of 0.1 and 1.36 MPa, and at superficial gas velocities of up to 9 cm/s with the Drakeol-10 oil as the liquid-phase medium and nitrogen as the gas-phase medium. Drakeol-10 oil has been used successfully by Air Products

Table 1
The physical properties of Drakeol-10 oil

T (°C)	20	100	175	200	265
Density [a] (g/cm^3)	0.849	0.806	-	0.7439	0.698
Viscosity [a] (cp)	38.13	3.38	-	0.892	0.55
Surface tension [b] (dynes/cm)	28.3	23.9	19.9	-	-

[a] Density and viscosity were measured by Huffman Lab, Inc.
[b] Surface tension was determined by Phoenix Chemical Lab, Inc.

and Chemicals, Inc. in a slurry-bubble-column-reactor as a starting slurry medium for the slurry phase methanol synthesis and Fischer–Tropsch synthesis. Drakeol-10 oil is a C_{16}–C_{38} saturated hydrocarbon liquid (food grade mineral oil) which includes straight chain and branched paraffins as well as naphthenic compounds. The physical properties of the Drakeol-10 oil are illustrated in Table 1.

3. Results and discussion

Fig. 3 illustrates the effect of superficial gas velocity (SGV) on the average gas holdup for experiments conducted at two different pressures (0.1 and 1.36 MPa) and a constant reactor temperature of 265°C. The average gas holdup was found to increase linearly with increasing superficial gas velocity. It increased from 0.11 at a gas velocity of 0.77 cm/s to 0.34 at a superficial gas velocity of 8.84 cm/s at 0.1 MPa. This is consistent with the results reported in the literature, i.e., the gas holdup increases with the gas velocity. Calderbank et al. [10] studied gas holdup in a 5-cm I.D. column using Krupp wax as the liquid medium at 265°C and 0.1 MPa. Gas holdups were found to vary linearly with gas velocity, with gas holdups reaching approximately 0.2 at a gas velocity of 5.5 cm/s. The result from this study is in good agreement with Calderbank's data [10]. The predicted gas holdup by using the general correlations reported by Bukur et al. [11] and Kuo [4] are also shown in Fig. 3. Bukur et al. [11] proposed a general correlation for the prediction of gas holdup.

$$\epsilon_g = 0.24\left(Fr_g^{0.28} Bo^{0.14}\right), \qquad (1)$$

where $Fr_g = u_g^2/(gD_c)$, $Bo = D_c^2 \varrho_l g/\sigma_l$.

Kuo [4] suggested a correlation for the gas holdup based on Mobil's 5-cm hot-flow bubble column.

$$\epsilon_g = 6.5 u_g^{1.1}. \qquad (2)$$

As can be seen in Fig. 3, the gas holdup predictions based on Bukur's [11] correlation is much lower than our experimental observations. While the prediction from Kuo's [4] correlation was lower for a lower gas velocity (≤ 4.26 cm/s) and higher for a higher gas velocity (≥ 4.26 cm/s). It can be seen that neither Kuo [4] nor Bukur et al. [11] correlations can predict the observations from this study. This is due to the fact that the gas holdup is sensitive to the physical properties of the dispersion media and the influence of liquid properties as well as the type of sparger used. Fig. 3 demonstrates that the increase of reactor pressure from 0.1 to 1.36 MPa has no significant effect on the observed average gas holdup. No effect or only a very small effect of pressure on gas holdup was observed by Kolbel et al. [12] and Deckwer et al. [6]. Kolbel et al. [12] found no pressure effect on the gas holdup up to 1.6 MPa for air–water system. Kuo's study [4] also shows

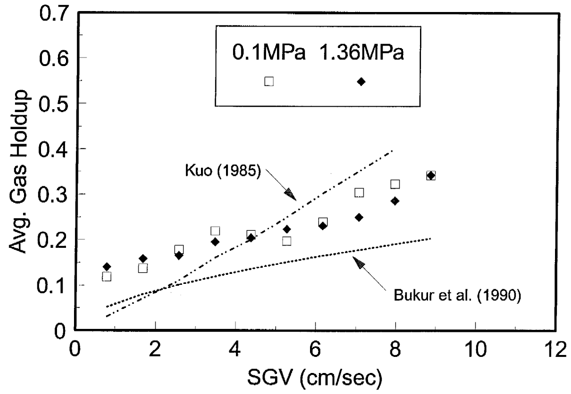

Fig. 3. Effects of pressure and superficial gas velocity on the average gas holdup at 265°C.

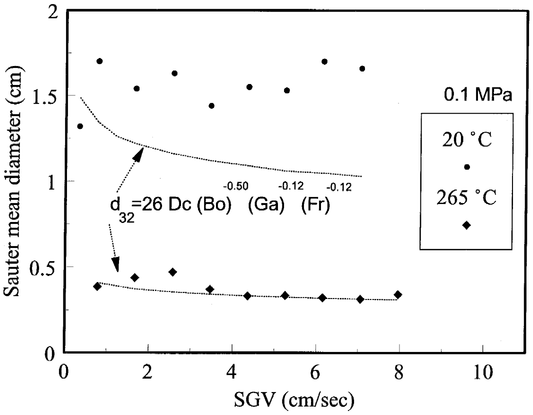

Fig. 4. Effects of temperature on the Sauter mean diameter at 0.1 MPa.

similar trends suggesting that the effect of pressure on the average gas holdup was insignificant. This is in good agreement with our observations. Idogawa et al. [13] reported the behavior of bubbles in a air–water system in a 5-cm-diameter column over a range of gas velocities from 0.5 to 5 cm/s and at pressures up to 15 MPa under ambient temperature. The gas holdup increases with increasing pressure under Idogawa's studies. This discrepancy in the effect of pressure on gas holdup is possibly due to the different systems and operating conditions utilized.

Fig. 4 shows the Sauter mean bubble diameters versus the superficial gas velocity at two different temperatures (20 and 265°C) at a constant pressure of 0.1 MPa ($d_{32} = (\Sigma n_i d_i^3)/(\Sigma n_i d_i^2)$). The bubble size decreased substantially when the temperature was increased from 20 to 265°C. It was about 1.5 cm at 20°C and decreased to approximately 4 mm as the temperature increases from 20 to 265°C. The physical properties of the Drakeol-10 oil as a function of temperature are illustrated in Table 1. The physical properties change significantly as the temperature increase to 265°C. These changes may be associated with the observed hydrodynamic parameters. This is consistent with the results from the literature, i.e., the lower the viscosity of the liquid medium, the higher the gas holdup. The bubbles are also larger at lower temperatures. The average bubble size is smaller because of the lower surface tension at the higher temperature [14]. The Sauter mean bubble diameter profiles in the bubble column were rather uniform at both temperatures (Fig. 4). Thus, the superficial gas velocity did not have any significant effects on the Sauter mean bubble diameter. Saxena et al. [15] reported that the bubble size may be independent of gas velocity in the 3.6 to 9.2 cm/s ranges for air–water system with a perforated plate distributor. Our result in Drakeol-10 oil/nitrogen system is in good agreement with Saxena et al. [15] observations. Akita and Yoshida [16] proposed the following correlation:

$$d_{32} = 26 D_c (\mathrm{Bo})^{-0.5} (\mathrm{Ga})^{-0.12} (\mathrm{Fr})^{-0.12}, \qquad (3)$$

where $\mathrm{Bo} = g D_c^2 \rho_l / \sigma$, $\mathrm{Ga} = g D_c^3 / \nu_l^2$, $\mathrm{Fr} = u_g / (g D_c)^{0.5}$ for the Sauter mean diameter in bubble columns with diameters up to 30 cm and with superficial gas velocities up to 7 cm/s. This correlation was used to calculate the bubble sizes in this study. As shown in Fig. 4 the predicted values at 265°C and 0.1 MPa are in good agreement with the experimental data. However, this correlation fails to predict the data collected at 20°C in this study.

Fig. 5 demonstrates the effect of pressure on the Sauter mean bubble diameter at 265°C. The bubble size decreases as the pressure increases.

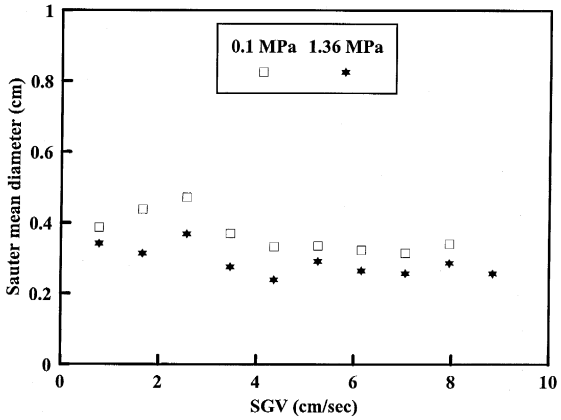

Fig. 5. Effects of pressure on the Sauter mean diameter at 265°C.

It was approximately 4 mm at 0.1 MPa; however, it decreases to approximately 3 mm as the pressure increases to 1.36 MPa. The Sauter mean diameter obtained at 265°C and 0.1 MPa was about 4 mm. This is consistent with the results reported in the literature, for example Bukur et al. [11] reported diameters of 4 to 5 mm for Mobil wax, Calderbank et al. [10] reported values of 3 to 5 mm for Krupp wax at similar reaction conditions. More recently O'Dowd et al. [7] obtained Sauter diameters in a reactor wax at 250 oC and 1.48 MPa. Their values were between 2.7 to 3.9 mm. It has been reported that the bubble size decreases with increasing pressure at a constant superficial gas velocity [17]. Idogawa et al. [13] also reported that the bubble diameters of the air–water system in a 5-cm-diameter column decreases with increasing pressure up to 15 MPa. Several factors that may affect the bubble size in bubble columns operated at elevated pressures are the formation, the coalescence and the breakup of the bubble. Firstly, the formation of the bubble, the increased pressure results in increased gas densities therefore the momentum of the gas leaving the distributor orifice is increased. The bubbles are forced to separate themselves from the distributor at an earlier stage of growth because of the upward force from the entering gas [18]. Thus, the bubble sizes leaving the distributor are reduced. The second factor is a reduced tendency of bubbles to coalesce. Pressure is likely to have some influence on it as there is a tendency for bubbles to minimize their volume to surface area ratio with increasing pressure [18]. Thirdly, the higher pressure influences the bubble breakup has been reported by Wilkinson and Dierendonck [17]. The decrease of bubble size with increasing pressure observed in our study is probably the combination all three factors.

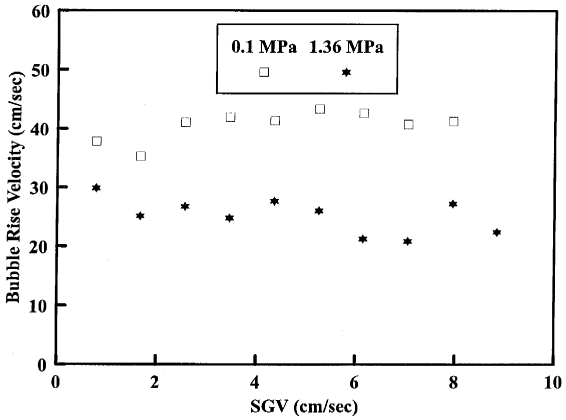

Fig. 6. Effects of pressure on the bubble rise velocity at 265°C.

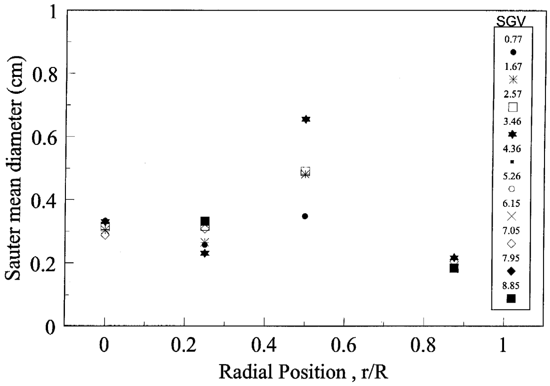

Fig. 7. Radial variation of sauter diameter at various gas velocities at 265°C, 1.36 MPa and 71 cm above the gas distributor.

Fig. 6 shows the effect of pressure on the average bubble rise velocity under a constant temperature of 265°C. The bubble rise velocity significantly decreases as the pressure increases. It was approximately 40 cm/s at 0.1 MPa; however, it decreased to approximately 25 cm/s as the pressure increased to 1.36 MPa. The bubble rise velocity profiles in the bubble column were rather uniform at both pressures. Therefore, the superficial gas velocity did not have any effect on the bubble rise velocity.

Fig. 7 shows the radial Sauter diameter profiles at a height of 71 cm above the gas distributor obtained on different superficial gas velocities under reactions conditions of 265°C and 1.36 MPa. Generally, radial Sauter diameter profiles were fairly uniform about 0.3 cm at a gas velocity of 0.77 cm/s, as expected since the flow is in the homogeneous bubbling regime at this velocity. As the gas superficial velocity increases, the flow becomes slightly non-uniform with approximately 0.3 cm at all radial positions in the column. It should be noted that at the radial position r/R of 0.5, the larger Sauter diameters were observed. It ranged from 0.48, 0.5 and 0.68 cm for superficial gas velocities of 1.67, 2.57 and 3.46 cm/s, respectively. At higher gas velocities, 7 cm/s and greater, the Sauter diameter profiles become more uniform all around 0.3 cm. The Sauter diameters were slightly less than 0.3 cm at the positions close to the wall. Fig. 8 illustrates the average bubble rise velocity collected at the same conditions as shown in Fig. 7. Generally, the trend of the average bubble rise velocity profiles were fairly similar for all superficial gas velocities. Those velocities were around 30 cm/s at the center of the bubble column. As the r/R increases from 0 to 0.875, the velocities decrease from 30 to around 25 cm/s. It should be noted that at the radial position of 0.5, higher bubble rise velocities were observed. They ranged from 38, 42 and 48 cm/s for superficial gas velocities of 1.67, 2.57 and 3.46 cm/s, respectively. The higher bubble rise velocities correspond to larger bubble diameters as shown in Fig. 7. Bukur et al. [11] reported the bubble rise velocity in the FT-300 wax–air system at 265°C and 0.1 MPa conditions. For small bubbles, the rise velocity increases dramatically with increasing size, and reaches around 15 cm/s for 2-mm bubbles. For large bubbles of size 10 mm, the rise velocity is about 25 cm/s. These numbers are in the same order of magnitude as found in this study: larger bubbles have higher rising velocities than smaller bubbles (Fig. 7 and Fig. 8). Kuo [4] also reported that the rise velocity (35 cm/s) of large bubbles is 4–7 times that of small bubbles (6 cm/s) in FT-200 wax system. The larger bubble sizes (Fig. 7) and faster rising bubble velocities (Fig. 8) for these bubbles in the region around $r/R = 0.5$ can also be explained by the flow characterization proposed by Fan [19]. Fan indicated that the flow in a bubble column under ambient conditions is characterized by a fast bubble flow region surrounding a central plume and a descending liquid flow region adjacent to the wall with vortices in between. Our results obtained under temperature and pressure are in good agreement with the flow patterns suggested by Fan [19].

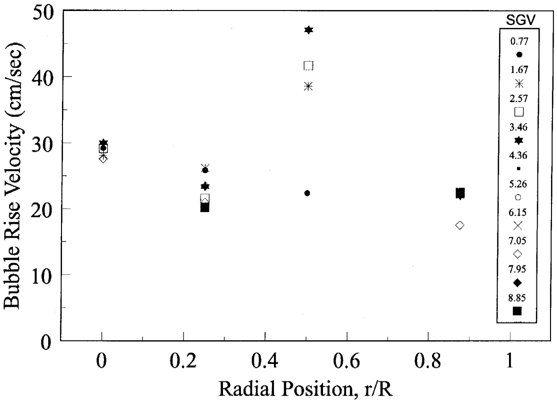

Fig. 8. Radial variation of bubble rise velocity at various gas velocities at 265°C, 1.36 MPa and 71 cm above the gas distributor.

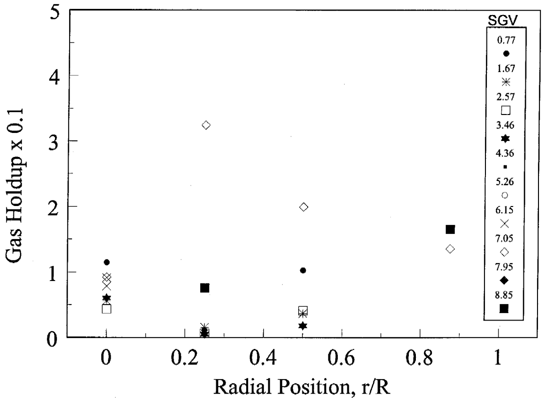

Fig. 9. Radial variation of gas holdup at various gas velocities at 265°C, 1.36 MPa and 71 cm above the gas distributor.

Fig. 9 shows the radial distribution of the gas holdup at 71 cm above the gas distributor obtained from different superficial gas velocities under reactions conditions of 265°C and 1.36 MPa. Generally, the local gas holdup increases with increasing superficial gas velocity. The local gas holdup profiles were fairly uniform for gas velocities less than 4.36 cm/s, as expected since the flow is in the homogeneous bubbling regime at those velocities. For gas superficial velocities above 7.05 cm/s, there was a signifi-

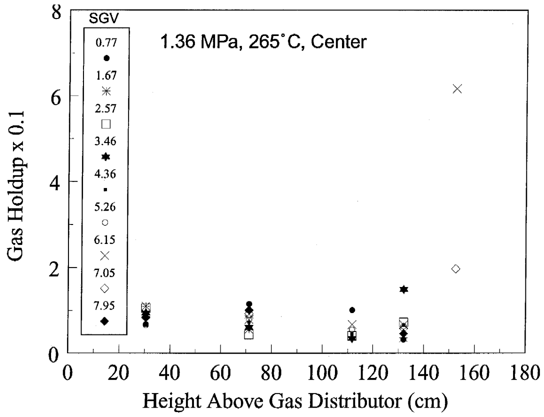

Fig. 10. Axial variation of gas holdup at various gas velocities.

cant change in the gas holdup for values of r/R between 0 and 0.875. The gas hold changes from 0.1 at the center of the column to 0.32 at $r/R=0.25$, then to 0.2 at $r/R=0.5$, and finally reduces to 0.15 at $r/R=0.875$.

Fig. 10 shows the effect of the gas superficial velocity on the axial gas holdup under conditions of 265°C and 1.36 MPa. For gas velocities of 4.36 cm/s or less, axial gas holdup profiles in the bubble column were uniform; however, at the gas velocity of 6.15 cm/s, there was a significant change in the gas holdup at a height of 130 cm above the gas distributor, the gas holdup increases from 0.1 to 0.6. At a higher gas velocity of 7.05 cm/s, the axial gas holdup also increases significantly in the column, particularly at 130 cm above the gas distributor. This increase in the gas holdup indicates the presence of a slug at the top of the reactor. Bukur el al. [11] have reported that the transition from the bubbly to the slug flow regime occurred between gas velocities of 4 and 6 cm/s. Likewise, in the large diameter column, the transition from the homogeneous bubbling regime to the churn-turbulent regime occurred between gas velocities of 4 and 6 cm/s. In the small diameter column, slugs start forming at 60 cm above the distributor. Our results obtained under pressurized conditions are in good agreement with Bukur's et al. [11] data.

4. Conclusions

The hydrodynamic parameters in a bubble column reactor were measured for the nitrogen/Drakeol-10 oil system at hot, pressurized conditions. The following results were obtained. The average gas holdup increases as the superficial gas velocity increases. The pressure in the reactor does not have any significant effect on the gas holdup. The Sauter mean bubble diameter decreases with increasing pressure and temperature at a constant superficial gas velocity. The bubble rise velocity significantly decreased as the pressure was increased. Akita and Yoshida's correlation provides a very good fit for Sauter mean diameters measured at 265°C and 0.1 MPa. Larger bubbles have higher rising velocities than smaller bubbles.

5. Nomenclature

Bo	Bond number
Fr_g	Froude number
Ga	Gallilei number
u_g	Superficial gas velocity
g	Gravity constant
D_c	Column diameter
d_i	Bubble diameter
d_{32}	Sauter mean diameter
n_i	Number of bubbles
Greek letters	
ν_l	Kinematic viscosity of liquid
ϵ_g	Gas Holdup
ρ_l	Density of liquid
σ_l	Surface tension

6. Disclaimer

Reference in this report to any specific commercial product, process, or service is to facilitate understanding and does not necessarily imply its endorsement or favoring by the United States Department of Energy.

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