INTRODUCTION

A knowledge of the hydrodynamics of bubble columns will be used in developing a clear rationale for the design and scaleup of the Solvent Refined Coal process (SRC-II) reactor. The scaleup of the SRC-II process calls for taking limited experimental data obtained from two feet diameter reactors and smaller and extrapolating to diameters of ten feet or larger. While the SRC-II reactor involves a three phase system, a two phase study involving non-Newtonian fluids should provide important understanding of the hydrodynamics.

Use of the power law model will provide insight into the behavior of the fluids in liquefaction reactors. The behavior of the flow system in liquefaction reactors is complex but there are regions in which the heterogeneous fluid would behave as a purely viscous material.

Further the current flow maps do not properly describe the hydrodynamic behavior in the region in which direct coal liquefaction reactors are to be operated. These limitations in the current literature point out the importance of describing boundaries and flow patterns for liquefaction reactors. A better understanding will lead to a clear rationale for a scale up procedure useful to direct coal liquefaction reactors.

For efficient contacting between a coal slurry and a gas in coal liquefaction reactors, bubble columns are often used as there is no requirement for moving parts, only a small floor space is required, and large mass transfer area and large mass transfer coefficient exist. The resemblance between a liquid and a slurry is very valuable and allows one to study two-phase gas-liquid systems. Design calculations for gas-liquid systems are based upon information on holdup and flow-pattern of the fluid phases since results of past experiments described in the literature are given in this way.

The variation of gas hold-up with physical, flow, and geometric parameter has been given considerable attention recently. The parameters considered included superficial gas velocity, superficial liquid velocity, column diameter, liquid viscosity, liquid density, gas density, surface tension, type of distributor system, as well as others.

Currently there is a lack of holdup and flow map data for non-Newtonian fluids as applied to bubble columns. Since direct coal liquefaction reactors operate with the fluid behaving in a non-Newtonian manner over part of the axial distance, data of this type will contribute to the understanding of design and scaleup techniques for these bubble columns.

For a given gas-liquid system in a pipe, the flow patterns vary with changes in gas and liquid flow rates. The flow maps give a graphical representation of transition boundaries in a two dimensional coordinate system. Their applications are generally restricted to a particular system and geometry. Coal liquefaction reactors are expected to operate in the bubble or bubble-slug flow patterns. For scaleup purposes experimental bubble column reactors should be operated in the same flow pattern as direct coal liquefaction reactors.

Theoretical analysis of flow patterns begins with attempts to describe the transition boundaries with appropriate equations for viscous and non-Newtonian liquids. These transition boundaries are dependent on liquid properties such as surface tension, viscous behavior and density. Evaluation of these properties for the fluids being used in the experiments were made. The transition boundaries and holdup expressions can be related to the coal liquefaction reactor to predict gas holdup, flow pattern, interfacial area and backmixing coefficient. An exploration of coordinate systems for presentation of the flow maps was undertaken.

Experiments were performed to measure gas holdup and to analyze bubble sizes, shapes, and distribution in the 0.3366 m column. Radial distributions of these variables were determined in air-water and air-aqueous CMC solutions. Analysis and modelling using all experimental data was done to determine predictive equations for holdup and flow patterns especially in the bubble-slug flow pattern. The effect of gas distributor on gas holdup was studied with pore size, thickness and material of the distributor as the variables. Interfacial area was determined by physical methods by measuring gas holdup and bubble size. Holdup data was analyzed for axial variation using sieve and porous plate gas distributors. An entrance region of about 1m was observed in the column. Radial and axial distribution of bubble sizes was obtained in the bubble column by means of a borescope.

Gas holdup was measured using up to 2wt% CMC solution to determine the viscous non-Newtonian effect on gas holdup. Foam accumulation in the column resulted in experimental errors in gas holdup measurements.

Modelling was done for a Newtonian fluid in bubble-slug flow and for a non-Newtonian fluid in slug flow. The effect of column diameter was studied by performing the experiments in 6" and 13" bubble columns.

Both the use of non-Newtonian liquids in bubble column experiments and the theoretical and empirical modelling of flow pattern boundaries are new areas of inquiry and as such will lead to important contributions to the understanding of scaleup in bubble columns.

OBJECTIVES

There are four major objectives for this proposed study. These objectives are basic to the understanding needed to develop a rationale for scaleup in bubble columns. This understanding is the key to improving our scientific and technical knowledge of the fundamental process involved in complex two and three phase flows.

These objectives are:

1. to properly characterize two phase flow patterns in the region of interest that direct coal liquefaction reactors will be operated.
2. to characterize for viscous liquids, Newtonian and non-Newtonian, the flow pattern boundaries in the operating region of direct coal liquefaction reactors. The characterization would include both empirical and theoretical models.
3. to develop empirical expressions and models for the gas holdup in the flow patterns of interest. This objective would focus on non-Newtonian liquids that follow some elementary models for constitutive behavior.
4. to determine the variation of bubble size along and across a bubble column and its effect on interfacial area for non-Newtonian liquids in the flow patterns of interest.

MAJOR ACCOMPLISHMENTS

SUMMARY

A review of literature and reports indicate

- o a bubble column coal liquefaction reaction will operate at or near the bubble to bubble-slug transition
- o small diameter hydrodynamic experiments do not apply to large diameter columns
- o no good generalized flow map exists for vertical cocurrent upflow

Gas holdup measurements in bubble columns indicate

- o an entrance region of about one meter
- o no column diameter effect between 6 and 13 in.
- o use of porous plate gas distributors significantly increases gas holdup in the bubble pattern over sieve plate distributors
- o a maximum exists when using porous plates
- o holdup varies with the distributor plate material
- o hydrophobic gas distributor gives higher holdup than hydrophilic distributor
- o a decrease as apparent viscosity is increased
- o similar shapes when using non-Newtonian liquids (carboxy methyl cellulose) or water

Bubble diameter measurements and observations indicate

- o a unimodal distribution in the bubble pattern
- o a bimodal distribution in the bubble-slug pattern
- p a borescope can be used to determine small bubble diameters

Interfacial area was observed

- o to reach a maximum when porous plate gas distributors are used
- o to be predicted correctly in the bubble-slug pattern when a two term equation is used
- o to decrease as apparent viscosity is increased

Modelling and analysis of experiments indicate

- o there is a band of transition between the bubble and bubble-slug flow patterns
- o slug flow would not exist in coal liquefaction reactors
- o average column shear stress can be predicted in the slug pattern
- o gas holdup in the bubble pattern with porous plates is correlated with gas velocity, pore size and thickness.
- o gas holdup in non-Newtonian systems in the slug pattern can be predicted with reasonable accuracy.
- o interfacial area can be correlated as a function of gas holdup in bubble and bubble-slug pattern for both Newtonian and non-Newtonian systems

Bubble frequency measurements in bubble columns indicate

- o large bubble frequency in the slug pattern is relatively constant over a wide range of apparent viscosity.

DISCUSSION

It has been observed that two phase flow occurs in one of several different patterns. Which pattern a pair of fluids are flowing in depends on the respective velocities and the physical properties of the fluids. For two phase concurrent vertical flow, six different patterns are possible but only two are important in direct coal liquefaction bubble columns. These include bubble or homogeneous flow and bubble-slug transitional flow or heterogeneous flow.

The bubble flow pattern is observed at low gas and liquid velocities. This pattern is characterized by an unimodal distribution of bubble sizes and bubbles which rise independently of one another. This type of flow is often termed homogeneous or pseudohomogeneous due to the uniform bubble size and the uniform distribution of bubbles in the column.

The bubble-slug pattern occurs when the gas velocity is increased to the point where the individual bubbles interact to a large extent. Bubble coalescence occurs in this pattern, forming a bimodal distribution of bubble sizes. The larger bubbles tend to rise in the center of the column, this causes the liquid to circulate in distinct cells inside the column. Backmixing is thus greatly increased in this pattern.

A 0.3366 m diameter bubble column has been constructed approximately 5 m high and a 0.1524 m diameter 5 m high bubble column has been modified for this work.

More than 20 gas distributors have been used in experiments performed in the 0.1524 m diameter bubble column trying to determine the effect of the distributor on the transition from bubble to bubble slug flow. The use of sieve plates clouds the transition from the bubble to the bubble-slug flow pattern. The variables studied included porosity, particle size, thickness, material and the holdup and velocity at the transition.

Since it appears that the coal liquefaction reactors will operate in the bubble-slug pattern, identification of gas holdup for each of the two major sizes of gas bubbles (bimodal distribution) has been accomplished. Dynamic gas disengagement experiments have been performed to help characterize this pattern by determining the velocity of both bubble sizes.

Seventeen gas distributors have been used in experiments using air and water flowing up cocurrently in the 0.1524 m diameter bubble column. The distributor variables studied included porosity, particle size, thickness and material. The holdup and velocity at the transition and their effect in the bubble and bubble-slug pattern and the transition between the two were measured. Direct coal liquefaction reactors will probably operate in the bubble-slug pattern. Identification of gas holdups for each of the two major sizes of gas bubbles (bimodal distribution) has been completed. Dynamic gas disengagement experiments have been performed to help characterize this pattern by determining the velocity of both bubble sizes.

A set of experiments have been completed using a non-Newtonian liquid, aqueous carboxy methyl cellulose solution to study gas holdup in non-Newtonian liquid systems and describe flow pattern changes due to these liquid solutions. Flow pattern transitions have been examined as well as bubble size and bubble distribution. Development of a model of the bubble-slug pattern to predict the important parameters of this flow pattern is underway.

Distinctive differences in gas holdup occur due to the use of different gas distributors. In the bubble flow pattern higher gas holdup occurs when using porous plates than sieve plates. In the bubble-slug flow pattern the gas holdups are essentially the same for both types of gas distributors. Holdup and gas velocity varied at the transition between bubble and bubble slug flow for different porous plate gas distributors.

Gas holdup using non-Newtonian liquids decreased over those of water in the bubble flow pattern. This decrease took place for concentration of CMC in water greater than one wt%. In the bubble slug pattern the holdup values were about the same as water even though the CMC solution had an apparent viscosity about an order of magnitude greater than water. A reduction in turbulence due to the increase in apparent viscosity may be the reason for the decline.

A series of dynamic gas disengagement experiments have been performed using the six inch inside diameter column. The major objective of these experiments was to determine whether the distribution has any effect on the bubble rise velocities. No major difference was found when the data for a porous plate and a sieve plate was compared.

Dynamic gas disengagement (DGD) occurs when at a certain instant, the gas supply to the column is stopped. The dispersion level will decrease with time as the gas continues to escape. Use of the dynamic gas disengagement technique in bubble-slug flow allows determination of the large and small bubble holdups separately.

Holdup and flow pattern studies were performed in a 0.1524 m diameter bubble column for the air-water system using 17 different porous plate gas distributors. The porous plates studied included polyethylene, polypropylene, aluminum oxide and silicon carbide, of thicknesses 0.125 inch to 1 inch, and pore sizes 35 to 240 micrometers. The bubble column was operated in bubble and bubble-slug flow patterns. The bubble to bubble-slug transition occurred between 0.054 to 0.085m/s depending upon the gas distributor. The gas holdup at the transition ranged between 0.2 and 0.33 for different gas distributors. Holdup increased linearly with superficial gas velocity and dropped suddenly at this transition due to coalescence of bubbles.

In porous plate distributors holdup increased with increase in pore size in the bubble and bubble-slug flow patterns. In our range, the increase in holdup is about 15% in the bubble flow pattern and about 8% in the bubble-slug flow pattern. Distributor plates with hydrophobic characteristics gave higher holdup than with hydrophilic characteristics in the bubble flow pattern. The variation was about 11% in the bubble flow pattern and the bubble slug pattern variation was less than 5%.

Polypropylene with hydrophilic characteristics gave higher holdup than polyethylene with hydrophilic characteristics. The difference in the bubble flow pattern was about 11%. Holdup for the hydrophilic polypropylene plate was less than that of polyethylene with hydrophobic characteristics, probably because of different wetting characteristics of the distributor. A summary of these conclusions follows:

1. Porous plates can have significantly higher holdup in the bubble pattern than sieve plates.
2. Porous plates have about the same or slightly higher holdup in the bubble-slug pattern.
3. Thicker porous plates usually have higher holdup in the bubble pattern.
4. For porous plates gas holdup increases with increasing pore size in the bubble pattern.
5. Gas holdup varies with the material of construction of porous plates.

6. Hydrophobic porous plates give higher holdup than hydrophilic.

Gas holdup measurements for air and aqueous carboxy methyl cellulose solutions have been performed using the 13 inch acrylic column. The experimental data has been analyzed and is reported below. A 1/4 inch thick porous plate with 70 micrometer pore size made of polyethylene and an acrylic sieve plate with 1/8 inch diameter holes were used as gas distributors. Holdup was measured at different axial positions and compared to total holdup measurements by summing. The gas velocity range extended over the bubble and bubble-slug patterns.

Based on the results of this study a number of conclusions may be drawn about the effect of non-Newtonian liquids on two phase flow parameters:

1. Rheological properties can affect gas holdup in bubble columns. The way in which these properties affect gas holdup depends on the flow pattern the column is operating in. In the bubble flow pattern gas holdup increases with viscosity at a particular gas velocity. In the bubble-slug or heterogeneous flow pattern, gas holdup generally decreases with viscosity.
2. Liquid velocity affects gas holdup in the bubble flow pattern. Increasing the liquid velocity reduces the gas holdup at any particular gas velocity. This effect is accentuated by increased viscosity.
3. The bubble to bubble-slug transition is dependent on viscosity. The trnasition gas velocity decreases with increasing gas viscosity. There is no effect of liquid velocity on this transition however in the range studied.
4. The effect of alcohol on two-phase flow parameters was extreme. Holdup in the bubble flow pattern was very high, often

with peaks greater than 0.5. The bubble to bubble-slug transition occurred at higher gas velocities than in aqueous CMC solutions. There also appeared to be competing effects between alcohol and CMC concentrations.

Variation of bubble size along and across a bubble column type reactor and its effect on interfacial area when a non-Newtonian liquid is used has been determined. Air and CMC solutions flowing cocurrent up were studied in the 0.3366 m I.D. bubble column. Only the bubble and bubble-slug patterns were taken into consideration.

Gas holdup, bubble size, and specific interfacial area studies were made in a 0.3366 m inside diameter bubble column with air-carboxymethyl cellulose aqueous solution as the system. Experiments were performed with a fixed porous plate gas distributor by varying superficial gas velocity. The flow patterns of interest were bubble and bubble-slug patterns. Gas holdup data was obtained by the bed expansion method and bubble size distribution by taking photographs wih a borescope at different radial and axial positions.

Gas holdup decreased with carboxymethyl cellulose (CMC) concentration and exhibits a maximum with superficial gas velocity. This maximum also diminishes with CMC concentration.

Bubble size measured by the borescope is smaller near the walls and reaches its maximum at R/2. The bubble size increases when either CMC concentration or superficial gas velocity is increased. There is no substantial variation of bubble size in the axial direction for the lower portion of the column. A maximum for total interfacial area was found for all the solutions with the exception of the higher concentrations. This maximum was found in the bubble-slug pattern near the transition from bubble to bubble slug pattern. Operation under this condition is recommended.

Modelling and analysis of experiments has been completed for both Newtonian and non-Newtonian systems in bubble columns. A bubble slug model has been developed for air-water system in a bubble column. A plot of assumed large bubble void fraction values versus calculated values of the large bubble void fraction from the bubble slug model indicates a unique solution for this model.

A viscous slug flow model has been developed for 2wt% CMC solution in a 6 inch diameter column as it is possible for large diameter bubble columns to operate in the slug pattern at large liquid viscosities. This model can also calculate the average shear rate of the liquid phase in the column.

Gas holdup correlations have been obtained for Newtonian systems in bubble and bubble slug patterns. Gas holdup in bubble pattern is correlated with gas velocity, pore size and thickness for Newtonian system. Gas holdup is correlated with gas velocity in bubble, bubble-slug and slug patterns for non-Newtonian system (CMC).

BACK GROUND INFORMATION

A review of the literature has been made to broadly classify the existing flow patterns in vertical two-phase gas-liquid upflow, which can occur in a bubble column. Also gas holdup, bubble size, and interfacial area data previously observed in bubble column operations will be classified.

FLOW PATTERN

A study of the phase distribution and shapes of interfaces can be of great use in the overall understanding of two-phase flow. The particular flow pattern one observes depends on the flow rates, fluid properties and the tube size. Heat and mass transfer rates, momentum loss, rates of backmixing, and residence time distributions all vary greatly with flow pattern. In order to be able to analyze and predict the formation of a particular flow pattern or the transition from one to the other, it is important to classify the flow patterns. Given the existence of any one pattern, it is possible to model the flow so as to predict the important process design parameters.

The five basic flow patterns for upflow observed by major researchers (59,153,162,164) have been designated as bubble flow, finely dispersed bubble flow, slug flow, churn flow and annular flow. The above mentioned flow pattern transitions are generally observed at constant low liquid rate with increasing gas rate. Finely dispersed bubble flow is observed at high liquid rates.

A flow map is important for use in predicting the flow pattern that exists at a given set of gas and liquid flow rates and physical properties of the components used. The flow maps give a graphical representation of transition boundaries in a two-dimensional coordinate system. The map coordinates used have been both dimensional and dimensionless. Flow maps have limited use when dimensional coordinates are used since the effect of certain parameter changes, typically diameter and viscosity changes, cannot be accounted for easily. Representation in dimensionless groups to overcome this limitation is itself a problem as there is a limited theoretical basis on which to select groups. In fact more than two are probably required to characterize each transition boundary. Thus the only solution is to use physically meaningful coordinates, such as combinations of dimensionless numbers applied in a restricted sense. The applications are generally restricted to a particular system and geometry.

Most flow maps have been drawn from experimental observations (59,162). Classification was made with little basis on theory. Generalizations have been difficult to make. Experiments were also performed with small tube diameters, which are not applicable for scale-up. In most cases no physical mechanisms were presented to explain why such transitions occur. Taitel et al (162) along with Govier and Aziz (153) (to some extent) have analyzed the physical mechanisms that occur at each transition. The transitions have been mathematically modelled and the resulting equations have taken into account the influence of fluid properties, pipe sizes and flow rates.

The various flow maps obtained have been described by many authors. Griffith and Wallis (154) used algebraic combination of gas and liquid input flow rates and pipe diameters to characterize flow transitions. The map does not distinguish between slug and froth flow. The flow maps so produced cannot be applied to systems other than air-water. Ellis (148) studied flow transitions using tube diameters from 1 to 30 cm. At higher liquid velocities no slug flow was observed. For column diameters larger than 12.5 cms transitions from bubble to slug is independent of column size. An extensive study of two phase vertical upflow has been made by Govier and Aziz (153). They correlated flow transitions with changes in pressure drops, holdups, and superficial gas velocity for the air-water system. Oshinowo and Charles (160) have studied glycerine, butanol-air systems and presented quite accurate flow maps, but gave no provision for a diameter effect. Taitel, Bornea, and Dukler (162) have considered flow maps modelling based on the physical phenomenon of transitions. The coordinates used are superficial velocities of gas and liquid and can be determined for any two phase system for which the properties are known.

Golan and Stenning (149) in the process of modifying the flow maps of Griffith and Wallis (154) introduced transition line between slug and froth flow. They experimented with air-water system in 1-1.5" diameter pipes and generated a "complete vertical upward flow map." Ellis (148) experimented with tubes varying from 0.4 to 12" in diameter, for air and various liquids, with the aim of correlating holdup. Based on this correlation, various flow pattern transitions can be drawn and so the correlation can be considered as a flow map. Very recently Taitel et. al. (162) have come up with a flow map based on models developed to predict transition boundaries between the four basic flow patterns: bubble, slug, churn and annular. Each transition is shown to depend upon the flow rate pair, fluid properties and the pipe size, but the nature of the dependence is different, for each of them, as the mechanisms that control them are not the same.

Gould et. al. (150) used a modified Dun and Ros map, but the selection of coordinates was empirical. Cichy et. al. (22) have drawn a flow pattern map on the Baker's coordinates, using data of Govier et.

al., and taking into account fluid properties and pipe diameter. As reported by Choe (145), Alves has redrawn the Cichy's flow map, the transition to mist flow has been added based on data from Collier and Hewitt (146).

Oshinowo and Charles (160) proposed a correlation similar to that of Griffith and Wallis (154), consisting of delivered gas volume fraction and the mixture Froude number.

The holdup is assumed to be a function of Froude number and the flow pattern. With the Froude number a property modifying group was included to account for different fluid systems (61). Spedding et al., (161) have tried to use similar coordinates and have drawn flow maps for pipes at different inclinations ranging from -90° (vertical down flow) to 90° (vertical upflow). The fourth root of Froude number was plotted against the ratio of liquid to gas flux.

All these studies discussed so far indicate a need for more experimentation and analysis of different systems for a better understanding of the rationale used in design and scale-up. The flow maps available in the literature are generally applicable to a particular system and geometry.

Two phase flow phenomena involving non-Newtonian liquids will differ from Newtonian liquids, possibly even qualitatively. Newtonian and non-Newtonian liquids produce different flow patterns. Newtonian liquids tend to produce circulating patterns, whereas non-Newtonian liquids produce stream-line deflection around gas slugs. Since the physical properties and liquid velocity distributions affect the holdup and flow pattern, a study involving non-Newtonian liquids is required on fundamental grounds. These fundamental observations should lead to improved design methods for reactors. Further bubble breakup in high viscosity and non-Newtonian fluids has not been clarified (11).

HOLD-UP

The variation of gas hold-up with physical, flow, and geometric parameter has been given considerable attention recently. The parameters considered included superficial gas velocity, superficial liquid velocity, column diameter, liquid viscosity, liquid density, gas density, surface tension, type of distributor system, as well as others.

It has been generally reported that the superficial gas velocity has a strong influence on holdup, increasing gas velocity increases gas holdup. Hughmark (67) reported that small columns (up to about 5 cm diameter) show a markedly higher gas hold-up at low gas velocities because of the absence of eddies. He also observed that gas holdup varies linearly for gas velocities up to 2.75 cm/sec. The liquid velocity in industrial bubble columns is very low as compared to gas velocity. Most experiments have been performed with stagnant liquid conditions, as it was observed that at very low liquid velocities (up to 2 cm/sec.) the liquid velocity had no effect on gas holdup (111). For high liquid velocity, gas holdup decreases with increasing superficial liquid velocity.

There is an uncertainty about the effect of column diameter on gas phase holdup. Fair et al. (35) analyzed experimental data obtained from 2.5, 5, 45 and 105 cm columns, using the same type of gas distributor, and showed that no appreciable change in gas holdup occurred. Hughmark (67) reported wall effects increase gas holdup at diameters up to 7.5 cm and that for diameters greater than 7.5 cm holdup is independent of column diameter. Kastanek et al. (74,76) determined that gas holdup increases with increasing column diameter from data obtained from

5,9,15,30 and 100 cm diameter columns and the effect was more significant at higher gas velocities. Research conducted with a commercial-scale (550 cm diameter) column (81), and compared to past research results (165) showed that the column diameter has little influence on average gas holdup if the column has as large a diameter as 550 cm.

Experimental results reported by Eissa et al. (34), showed that increasing the viscosity from 1 to about 11 cp is accompanied by increasing gas holdup, reaching a maximum at about 3 cp. Above 3 cp the gas holdup decreases sharply at higher gas velocities. From about 11 to 39 cp there is an almost constant but slow rate of gas holdup decrease with gas velocity. The behavior was explained in terms of hindered gas bubble motion in viscous liquids. Experiments performed by Javdani et al. (69) observed that for 700% change in viscosity (20 to 140 cp.), there was a 20% change in holdup. Holdup increased with increasing density of gas phase from data reported using helium and nitrogen (69). Increasing the surface tension of the continuous liquid phase is accompanied by a decrease of gas holdup (34).

Until recently holdup data was compared and analyzed for different systems but without considering the different geometries of the distributor. The gas holdup is highly sensitive to the gas distribution system. A comparison of three types of distributor systems, fritted glass, porous plate and orifice sparger, clearly indicated that fritted glass and porous plate distributors gave higher holdup than orifice type the rank being fritted, porous and orifice (80).

Godbole et al (48) found that holdup is lower for larger column diameters and also decreases with increasing distributor plate hole diameter. They gave a correlation for a broad range of viscosities in Newtonian liquids:

$$EG = 0.319\ VGS^{0.476} \mu_L^{.058}$$

and said that the equation fits most of the data with a 2.5% error while Akita-Yoshida correlation (1) gives 11% error.

Many authors (35,39,61,67,75,80,81) agree with the conclusions from Akita-Yoshida (1). Others sometimes partially agree with some of their conclusions (3,127,163). Most investigators (34,48,76,78,90,94) disagree with their finding. Kastanek et al (76) disagree with the common idea that knowing holdup data for at least 300mm. I.D. column diameter is enough for scaling up (112) to larger diameters. They established a correlation after testing diameters from 50 to 1000 mm using air and water given by

$$EG = \dot{\Upsilon}\ [(k+D)][VGS^{4/5}/(2VGS+20)^{7/15}]$$

where $\dot{\Upsilon}$ and k are constants that depend on the system, (for water-air: $\dot{\Upsilon}$ =0.1925, k = 45.6). They did not find any effect of liquid height on holdup.

Except for teflon porous plates, the material used for construction of porous plates does not have an appreciable effect on holdup. The unwettability of teflon seems to give larger bubbles resulting in a lower holdup (43).

Very good agreement exists in investigations involving electrolyte solutions (1,30,61). Researchers have found that the holdup in these solutions is slightly larger than in non-electrolytes due mainly to the electrostatic potential at the gas-liquid interface.

The affect of non-Newtonian behavior of liquids on two phase flow parameters recently has become of interest. As far back as 1964 however, this area has been studied.

Researchers (5,10,16,19,29,35) measured holdup in bubble columns containing non-Newtonian fluids. The most popular fluids used in these studies were solutions of carboxymethyl cellulose (CMC) in water. The types of gas distributors used were varied: Nakanoh et al (93) used a single orifice sparger in a 14.55cm column. Bucholz et al (19) measured holdup in a multistage column, stages were separated by perforated plates.

Working with CMC solutions Schumpe and Deckwer (114) found that the gas holdup increases with CMC concentrations up to a concentration of 0.8% at low gas velocities. This is in disagreement with results from Franz et al (38) and Buchholz et al (19). The last two researchers also found holdup values lower for CMC solutions than for water at low gas velocities. Schumpe and Deckwer (114) recognized that gas holdup decreases with CMC concentration in the slug pattern.

There is some agreement on the presence of a maximum holdup when CMC solutions are tested with porous plates as gas distributors (32,48,114). This maximum, similar to other investigations (29,114), corresponds to the transition from homogeneous to heterogeneous patterns and is present at gas velocities near 1 cm/s. Also this maximum diminishes when CMC concentration is increased. When lower values of holdup are present using perforated plates (39), the holdup increases as the hole diameter is reduced.

For CMC concentrations higher than 0.8%, using a porous plate, Schumpe and Deckwer (115) suggest the following equation to be used only in the bubble flow pattern:

$$EG = 9.08 \times 10^{-2}\ VGS^{0.65}$$

and with a perforated plate:

$$EG = 2.58 \times 10^{-2}\ VGS^{0.876}$$

and in the slug pattern where the holdup does not depend on the type of distributor (39):

$$EG = 3.22 \times 10^{-2}\ VGS^{0.674}$$

and for diameters smaller than 10cm:

$$EG = 4.04 \times 10^{-2}\ VGS^{0.627}$$

This agrees with earlier conclusions about the dependence of holdup on column diameter (34,90,119).

Obviously, in the slug flow pattern the holdup does not depend on the viscosity of the liquid phase, so knowing this characteristic, Deckwer et al (32) correlated data in CMC solutions for superficial gas velocity above 2 cm/s:

$$EG = 0.0265\ VGS^{0.82}$$

where almost no difference is present using different distributors.

Godbole et al (48) did some experiments varying the apparent viscosity of CMC solutions from 0.018 to 0.23 Pa-s and correlated the data with an equation that accounts for viscous effects:

$$EG = 0.225\ VGS^{0.532}\mu_{APP}^{-0.146}$$

With this equation they predict data with only a 5% error. Using results from Schumpe and Deckwer for different column diameters (115) they correlate the overall data with:

$$EG = 0.239\ VGS^{0.634}\ D^{-0.50}$$

BUBBLE DYNAMICS

Bubble size and bubble size distribution are important parameters that effect the gas holdup, interfacial area and mass transfer coefficient.

Kumar et al (80) and Bhavaraju et al (11) have found that bubble diameter depends on the specific gas-liquid system and its properties with respect to coalescence. Coalescence is significantly influenced by the physical properties of the liquid. Calderbank (20) has found that bubble breakup is due to disturbances at the interface caused by external factors. Kozo Koide et al (82) found that bubbles generated from the porous plate are small and have an equal size, and wide size distribution, is observed when coalescence occurs.

Houghton et al (65) have found that the average pore size depends upon the physical properties of the liquid used, in particular the surface tension. They found that the lower the surface tension of the liquid the lower the average pore size for the same porous plate. They also have found that the bed density $\rho_B = \frac{\rho_L h_L}{h}$ is a function of both bubble size and the number of bubbles per unit volume of bed. For low viscous liquids (0.5-1.3cp) the bubble shape is insensitive to viscosity. Bubble size increases with surface tension, though not proportionally. The rise velocity of bubbles is given by

$$U_B = U_G \left(\rho_L/\rho_L - \rho_B \right)$$

The principal problem is how to approximate the bubbles shape to an average diameter for a hypothetical bubble. Many authors have presented good approximations (2,18,27,52,129) of bubble diameter. For example, Davies and Taylor (27) attempted to give a good and realistic equation for the approximation of spherical caps to spheres taking into account the radius of curvature of the caps.

$$Rc = 2.3\ Rb$$

Treybal (128) presents some empirical correlations for estimating average bubble diameters and concludes that to obtain very good estimates in air-water the following equation is the best:

$$d = \frac{2.344 \times 10^{-2}}{(VL/1-EG)^{0.67}}$$

Houghton et al (65) measured the major and minor axis of oblate spheroids in aqueous solutions with air and found the axis related by b=1.2a and recommended the use of the equivalent diameter of the spherical bubble for the same volume as the oblate ($D = (ab^2)^{1/3}$) so

$$De = 1.13\ a$$

Where 'a' is the minor axis of the oblate spheroid.

But in the most important region of two phase flow, the bubbles have a broad variety of shapes and it is important to take into account the distribution of them in order to end up with the best estimate of interfacial area. Akita and Yoshida (2) proposed to eliminate bubbles smaller than 0.8mm. Their contribution to holdup or interfacial area (38,39) can be neglected. The bubbles that are not spherical can be approximated by an oblate spheroid resulting in good agreement with Houghton et al (65). They found the mean volume-surface diameters followed a geometrical distribution function and gave a good estimate of the volume-surface diameter:

$$\frac{dvs}{do} = 1.88\ (VO/\sqrt{gDo}\ )^{1/3}$$

They developed good correlation for a broad variety of liquids in agreement with most of authors (67,76).

$$\frac{dvs}{D} = 26\ N_{BO}^{-0.5}\ (N_{Ga}\ N_{Fr})^{-0.12}$$

They found the mean diameter dVs decreased when column diameter was increased. With the help of the Akita-Yoshida (1) holdup relation, they found a correlation for interfacial area when holdup is below 0.14:

$$aD = 1/3\ NBo^{0.5}\ NGa^{0.1}\ EG^{1.13}$$

It is important to remark that although all their experiments were done with a sparger as gas distributor, they recommend this equation for perforated plates too.

Particularly important are the mean diameter analysis of Franz et al (38) and Ueyama et al (163). For the former, the use of the gas dynamic disengagement method (48) led to the inclusion of two main bubble sizes, one relatively large and another very small, (approximately 0.1mm) (48); and the calculation of the Sauter mean diameter (2,80) with the help of the holdup for both fractions (48):

$$dvs = \frac{f \sum nki \ dki^3 + \sum nGi \ dGi^3}{f \sum nki \ dki^2 + \sum nGi \ dGi^2}$$

Where $f = (Ek \sum nGi \ \ dGi^3)/(EG \sum nki \ \ dki^3)$ with the mean diameters behaving as a normal distribution. The Sauter diameter of small bubbles does not depend on gas velocity or gas distributor while the medium to large bubbles Sauter diameter increases with both.

Researchers (18,24,38,39,114,115) have studied the bubble size and bubble size distribution in bubble column reactors using non-Newtonian liquids. Bubble size and shape in a bubble column is influenced by the gas sparger and the liquid properties. Bubble size distribution is usually measured by photographic method and sulfite oxidation method. Sauter mean diameter is generally obtained from the equation

$$dvs = \frac{\sum ni \ dBi^3}{\sum ni \ dBi^2}$$

It was found that (34) at relatively low viscosities, drag forces are not large enough to cause bubble coalescence, and uniform distribution of bubbles gives rise to higher gas holdups. Higher drag forces promote coalescence which results in lower gas holdups. Buchholz et al (18) have measured bubble swarm velocities and found that small bubbles in swarm have higher rise velocities than single bubbles. They (18) also found that large single bubbles rise with high velocity due to the change of their shape caused by the swarm of small bubbles. They found unimodal, bimodal and trimodal distribution of bubble sizes using a porous plate sparger. The bubble diameter distribution changes significantly with increasing superficial gas velocity.

Godbole et al (48) have found two bubble sizes using perforated plate distributor. Nakonah and Yoshida (93) have determined bubble size distributions using photographic technique. They found very small bubbles of less than 1 mm in size and large bubbles using a single orifice sparger. Franz et al (38) have found that with increasing CMC concentration the Sauter mean diameter of intermediate to large bubbles increases. They (38) also found that the effect of concentration reduces with increasing perforated plate hole diameter.

Schumpe and Deckwer (114,115) have used photographic method and sulfite oxidation method to determine bubble size distributions. With sintered plate sparger they (114) found small and uniform bubbles at low gas velocities. With perforated plate sparger they found large bubbles with intensive coalescence. The Sauter diameter increased (115) with increasing gas velocity for the sintered plate and decreased for the perforated plate at low gas velocities. They (115) found that at low gas velocities the Sauter diameter is independent of the sparger.

Many authors conclude that the presence of even traces of surface active agents reduces coalescence of bubbles and increases their rise velocities. Koide et al (81) worked with and without surfactants and found smaller average bubble size in a narrow range when surfactants were used. Without the use of them the bubble size was bigger and a really broad size distribution was present. They correlate the bubble size with:

$$db(q\varrho/g\sigma)^{1/3} = 0.64\ (NFr/NWe^{1/2})^{0.1}$$

INTERFACIAL AREA

The gas-liquid interfacial area is an important design parameter which depends on the geometry of the bubble column reactor, the operating conditions and the physical properties of the liquid phase. Interfacial area is related to gas holdup and volume to surface mean bubble diameter by the equation.

$$a = 6EG/d_B$$

The effects of many variables on interfacial area in bubble columns have been studied by a large number of investigators.

Koide et al (81) found that the nature of the gas used in bubble columns does not give any significant effect on the behavior of bubbles. This simplifies the scope of any investigation as far as bubble size is concerned.

Knowing that between bubble and bubble-slug patterns, the interfacial area tend to reach a maximum some authors tend to characterize this as a transition. Specifically, Otake et al (99) found a ratio standard deviation/average bubble size near 0.15 in this region while far apart in the slug pattern the same ratio is approximately 0.4. Also Nakoryakon et al (94) found a maximum wall shear rate while increasing gas velocity and also that the maximum coincides with the transition from bubble to bubble-slug patterns.

Buchholz et al (18) have studied the effect of column diameter on specific interfacial area. They found that the diameter of the column influences the coalescence significantly, ie; the smaller the column diameter, the higher is the coalescence frequency and the smaller is the specific interfacial area.

Kastanek et al (74) presented a relatively good equation for calculating interfacial area in viscous solutions. Where glycerol aqueous solutions were the principal systems:

$$EG/a = de/4$$

de = equivalent diameter.

with 18% as the biggest error.

Yagi-Yoshida (139) conclude that the interfacial area and mass transfer coefficients in viscoelastic fluids seem to be smaller than in elastic fluids. This is due to the fact that in viscoelastic fluids large bubbles mingle with very fine bubbles whereas in elastic liquids they are relatively uniform in size.

Interfacial area for non-Newtonian liquids in bubble columns were measured by several researchers (39,114). Schumpe et al (114) measured interfacial area for CMC solutions by both chemical and photographic methods and found wide descrepancies between the two methods. The area found by the photographic method increased much faster with gas velocity than by the chemical method. Buchholz et al (18) measured interfacial area by the photographic method. They reported a trimodal distribution of bubble sizes under some conditions in CMC solutions.

It was found that the specific interfacial area increases with increasing gas velocity at all concentrations (114). Interfacial area decreases with increasing CMC concentration, and a correlation was given in terms of superficial gas velocity and apparent viscosity in the slug flow pattern (114).

$$a = 4.65 \times 10^{-2}\ VGS^{0.51} \mu_{eff}^{0.51}$$