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Interfacial Area Analysis of a Non-Newtonian Liquid and Air Flowing Cocurrently up in a Bubble Column

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INTERFACIAL AREA ANALYSIS OF A NON-NEWTONIAN LIQUID
AND AIR FLOWING COCURRENTLY UP IN A BUBBLE COLUMN

BY

CARLOS E. TRIGUEROS

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE
REQUIREMENT FOR THE DEGREE OF
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ABSTRACT

Gas holdup, bubble size and specific interfacial area studies were made in a 33cm 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 with a boroscope 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 boroscope 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.

ACKNOWLEDGMENT

The author would like to express his sincere gratitude to his major professor Dr. Harold N. Knickle for his guidance, help and encouragement throughout this research.

DEDICATION

I would like to dedicate this thesis to my wife, Xinia. Without her patience during the long hours I had to ignore her to pursue the research, and without her encouragement which helped me through difficult periods of the work and without her love, which made the work worthwhile, this thesis, which I hope is a contribution worthy of dedication to her, would never have been completed.

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NOMENCLATURE

- A = Distance between the axis of symmetry and the center of the vortex (eq. 12).
- CA* = Equilibrium liquid concentration.
- D = Column diameter.
- DA = Diffusivity of phase A.
- EG = GAS HOLDUP
- EG2 = Gas holdup or holdup due to large bubbles (eq.27).
- EG1 = Holdup due to small bubbles, also EGk
- EGt = Gas holdup at any time.
- K = Constant for eq. 5.
- NBo = Bond number ($g D^2 \rho_L / \sigma$).
- NFr = Froude number ($VG / \sqrt{g D}$).
- NGa = Galilei number ($g D^3 / \nu L^2$).
- NWe = Weber number ($\rho D VG^2 / \sigma$).
- NEo = Eotvos number ($g D e^2 \Delta \rho / \sigma$)
- QG = Gas volumetric flow.
- QL = Liquid volumetric flow.
- Rc = Radius of curvature.
- Reb = Reynolds for the bubble.
- Reh = Reynolds in the hole or sparger (eq.30).
- Vh = Velocity in the hole.
- VG = Superficial gas velocity.
- VL = Superficial liquid velocity.
- Vs = Slip velocity.
- Vb = Single bubble velocity.
- YL = Volumetric liquid flow fraction (eq.11).

z^* = Dimensionless length
 a = Specific interfacial area.
 db = diameter of the bubble.
 de = Equivalent diameter.
 dh = Orifice or hole diameter.
 DVs = Volume-surface mean diameter.
 e = Energy dissipation.
 g = Gravity constant.
 k = Reaction rate constant and constant for eq. 2.
 KL = Liquid mass transfer coefficient.
 m = Reaction order.
 n = Power law index, constant for eq. 2.
 r^* = Dimensionless radius.
 wo = Maximum vorticity (eq.12).
 ν = Kinematic viscosity.
 μ_{App} = Apparent viscosity.
 γ = Shear rate, and constant for eq. 2
 σ = Surface tension.
 δ = Porous size.
 η_0 = Apparent viscosity at $dV/dy=0$.
 η_∞ = Apparent viscosity at $dV/dy=\infty$.
 λ = Time constant in rheological characterization.
 $\Delta\rho$ = Liquid-Gas densities difference .
 S_i = Partial cross sectional area.
 α = Parameter in Kato's correlation $(= 1 - e^{-0.2 VG^2})$.

INTRODUCTION

Gas bubbles have an important role in many physical and chemical processes. In some operations the transfer of mass between gas bubbles and the contiguous liquid phase is the very essence of the operation (53).

The bubble column is widely used as a gas-liquid reactor because it can be easily designed for the use at high pressure on account of its simple structure, with no moving parts, and its easy fabrication (43).

Bubble column is of easy operating and maintenance owing to the absence of any mechanical parts (13), also bubble columns are not very costly and has appreciable mass transfer possibilities besides, it is well suited for relatively slow gas-liquid reactions with gases that are not very soluble (67). Its ratio performance/cost is often in a better position than its main rival, the mechanically agitated tank.

In practically all the gas-liquid systems, the liquid phase mass transfer resistance is strongly controlling (60,65). The use of bubble columns with their ability to handle high liquid rates is a field of application where the former problem is present.

Bubble columns are used in a large variety of gas-liquid reactions as in lime-slurry carbonation, chlorination of paper stock, and hydrogenation of vegetable oils (60).

In sewage treatment they are widely used where air has

to be passed through porous carbon diffusers (28) into sewage. Also recently, bubble columns gained more importance in the area of biotechnology, especially in fermentation and waste water treatment, because they provide favorable mixing and mass transfer properties at low shear stresses (13).

Perhaps, bubble columns are mostly used in coal liquefaction processes which deal with elevated temperatures and pressures. Coal is pulverized to fine size and dissolved in a hydrogen donor solvent. A coal slurry can be treated as a pseudo homogeneous mixture (16).

The feature common to the above applications is the utilization of a non-Newtonian liquid as the working medium.

This is the basis of interest for the present study.

In the design of bubble columns there are many variables that are important : flow pattern, bubble size, bubble size distribution, gas holdup, mass transfer coefficient, interfacial area, column diameter, backmixing, and vaporization of the liquid phase (57). Usually these variables are related to each other and the variation of one could affect the whole design.

The bubble and bubble-slug patterns are of interest because there is a maximum in the interfacial area at their transition (19). Interfacial area is not the only parameter used for scale up in large columns. Backmixing takes place and is detrimental for overall performance. This can be avoided with the inclusion of baffles in the column. For example, a bubble column with baffles and

agitation gives the same holdup values as in stirred-agitated vessels, that as will be explained latter will reflect in an increase in the interfacial area.

In scale up procedures many suggestions have been presented (10.,34,39). Kastanek (37) found a very good way to scale up bubble columns based on constant values of rate of energy dissipation. This appears to be a reasonable compromise for the majority of practical situations. He also related the energy dissipation to the interfacial area and holdup, the most important factors in the design.

Akita and Yoshida (2) found that a square column gives the same performance as a round column with the diameter equal to the side of the square.

OBJECTIVES

The objective of this study is to determine the 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. Air and CMC solutions flowing cocurrent up are studied in a 33 cm I.D. bubble column. Only the bubble and bubble-slug patterns are taken into consideration.

LITERATURE REVIEW

FLOW PATTERNS :

The flow in two phase mixtures may not be characterized merely as laminar or as a combination of laminar and turbulent flow, but the relative quantities and the distribution of the phase must be considered (23,24). However, the problem in two phase flow is that there is a lack of agreement in the description and classification of the flow patterns where the subjectivity of the observer is also variable (63). It is also necessary to know that maps prepared from data for one pipe size and fluids properties are not necessarily valid for other sizes or properties.

A good attempt at characterizing flow patterns in bubble columns was made by Govier et al (23) consisting of a graphic presentation of pressure drop vs. holdup at constant liquid velocity. This graph has the characteristic of two minima and a maximum and defines four different flow patterns. Pattern I ranges from zero gas velocity to the first minimum, pattern II is between this minimum and the maximum, pattern III is between the maximum and the second minimum and the last pattern beyond this last minimum. With visual observations on bubbles shape, they found similar divisions. These patterns are called : bubble, slug, froth, and annular mist. Their correlated data applied only to water-air systems using small diameter columns.

Taitel et al (63) have described flow patterns called bubble, slug, churn and annular.

A summary of the description of these patterns is given as follows :

Bubble flow :

The gas phase is approximately uniformly distributed in the form of discrete bubbles in a continuous liquid phase.

Slug flow :

Most of the gas is located in large spherical cap shaped bubbles which in small bubble columns occupy almost the whole diameter (Taylor bubbles).

Churn flow :

In this pattern the large bubbles become narrower and their shape is distorted and the pattern is chaotic, frothy and disordered. The continuity of the liquid is destroyed by a high local gas concentration.

Annular flow :

This pattern is characterized by the continuity of the gas phase along the pipe in the core. The liquid phase moves upwards partially as wavy liquid film and partially in the form of drops entrained in the gas core.

Subjectivity is usually inherent in visual classifications. Taitel et al (63) used equations to generalize the prediction of the transition boundaries.

Lately, the most common maps for two phase flow classification includes six different patterns (41) :

bubble flow, finely dispersed bubble flow, bubble-slug flow, slug flow, churn-slug flow and annular flow. As can be seen from this classification the patterns are roughly described by former definitions and can be graphically observed in figure 1.

Again, the main problem is how to characterize the transition between patterns. Kamat and Knickle (35) have characterized the transition from bubble to bubble-slug by :

$$V_L = 2.2 V_G - 1.128 (g \Delta \rho \sigma / \rho_L)^{1/4} \quad (1)$$

For the bubble-slug to slug transition Knickle and Kirpekar (41) suggested the following equation :

$$V_L = 1.79 V_G - 0.286 (g D \Delta \rho / \rho_L)^{1/2} \quad (2)$$

Equations (1) and (2) are related to most interesting region as far as interfacial area is concerned. More equations defining transitions from one pattern to another have been developed by some authors (35,41,63).

Some other contributions for description and definition of two phase flow patterns were given by many authors (10,56,67).

Among those attempts describing flow patterns, one of

Figure 1. Flow Patterns In Gas - Liquid Vertical Upflow

particular interest because its use in non-Newtonian liquids was correlated by Schumpe and Deckwer (56). They correlated data from a 14cm. I.D. bubble column for a sintered plate gas distributor using CMC solutions and presented a map in which VG is plotted against the apparent viscosity with all patterns represented.

NON-NEWTONIAN BEHAVIOUR :

The Newtonian behaviour in a fluid is attributed to a linear relationship between shear stress (γ) and shear rate ($\dot{\gamma}$ or dv/dy). Obviously, in the case of non-Newtonian liquid, such a linear relationship does not exist.

Included as non-Newtonian fluids are time dependent, time independent, and viscoelastic fluids (61). The time dependent fluids are those for which the rate of shear at a given point is solely dependent upon the instantaneous shear stress at that point. The time dependent fluids are usually classified into two groups : thixotropic and rheopectic fluids, depending upon whether the shear stress decreases or increases with time at a given shear rate and constant temperature (61). The behaviour of those fluids is presented in figure 2.

As can be seen from figure 2, the thixotropic fluids exhibit reversible decrease in shear stress with time at a constant rate of shear and fixed temperature. Rheopectic fluids which are relatively rare in occurrence exhibit a reversible increase in shear stress with time at a constant rate of shear under isothermal conditions.

The viscoelastic fluids are those that show partial elastic recovery upon the removal of a deforming shear stress. Such materials have properties of both fluid and elastic solids (61).

The time independent fluids are those for which the shear rate is a function of both the magnitude and the

FIGURE 2
NON-NEWTONIAN TIME
DEPENDENT BEHAVIOR

duration of shear and possibly of the time lapse between consecutive applications of shear stress (61). These are divided into three subgroups : fluids with a yield stress, pseudoplastics, and dilatants.

The physical behaviour of fluids with a yield stress is usually explained in terms of an internal structure in three dimensions which is capable of preventing movement for values of shear stress less than the yield value. The dilatant fluids observe an increase in apparent viscosity with increasing shear rate. The majority of non-Newtonian fluids, the pseudoplastic, are characterized by linearity between $\mathcal{T}$ and dV/dy at very low and very high shear rates (61). These different behaviours are presented in figure 3.

Pseudoplastic fluids are common non-Newtonian fluids and are of great importance in industrial applications (11,16). Frequently, the pseudoplastic behaviour of fluids is represented by the Power law or Ostwald-deWaele model (61) which relates the shear stress to shear rate as follows:

$$\mathcal{T} = k (dV / dy)^n \quad (3)$$

where k is a measure of the consistency of the fluid and the higher the k , the more viscous the fluid (47). The value n is a measure of the degree of non-Newtonian behaviour, and the greater its departure from unity toward zero, the more

FIGURE 3 NON-NEWTONIAN FLOW BEHAVIOR

pronounced are the non-Newtonian properties of the fluid (6) .

There are other important models as Ellis, De Haven, Prandtl-Eyring, etc. (61) that will not be considered here.

Following the definition of viscosity for Newtonian behaviour the non-Newtonian shear stress can be written as:

$$\tau = \mu_{app} (dv/dy) \quad (4)$$

Using the Power law model given by equation 3 we can solve for the apparent viscosity (61) by combining equations (3) and (4), and get

$$\mu_{app} = k (dv/dy)^{n-1} \quad (5)$$

The index n is always less than unity for pseudoplastic fluids, it is deduced that the apparent viscosity decreases with increasing shear rate.

CMC solutions follow pseudoplastic behaviour and can be described by the Power law model (17) although Shima and Tsujino (59) recommend the model :

$$\eta - \eta_0 = \frac{\eta_0 - \eta_\infty}{(1 + \lambda |dv/dy|)^n} \quad (6)$$

where λ is a time constant, n a material constant and η_0, η_∞ the apparent viscosities at zero and infinite dv/dy respectively.

Furthermore, Nishikawa et al (51) working with CMC solutions in two phase systems, recommended the shear rate calculation using the following equations :

$$\text{for } VG \geq 4 \text{ cm/s} \quad dv/dy = 50 \text{ VG} \quad (7)$$

$$\text{and for } VG \leq 4 \text{ cm/s} \quad dv/dy = 100 \text{ VG}^{1/2} \quad (8)$$

These equations are in agreement with other experiments (56).

When behaviour of polymer solutions or specifically CMC solutions are compared, the same rheological properties are not always reproducible since the polymers vary somewhat with degrees of polymerization and degradation (49).

GAS HOLDUP

Gas holdup is an important property for design purposes because of its direct influence on column size (65) and because it is related to the gas liquid surface area and hence to mass transfer (30). In most instances the holdup phenomenon is measured over a cross sectional area or a finite system volume (24). The total volume fraction of gas phase in a system is given by

$$EG = \frac{QG}{QG+QL} \quad (9)$$

or in terms of volumes

$$EG = \frac{VG}{VG+VL} \quad (10)$$

These equations do not represent local behaviour along the column (22).

VARIABLES IN HOLDUP :

The gas holdup in bubble columns is primarily dependent on gas velocity, liquid velocity, surface tension, gas and liquid viscosities, temperature and pressure of the system, roughness of the column, diameter of the column, gas distributor and liquid depth (23). However, due to the

slight differences found in the results when some of these variables are varied, many investigators tend to neglect most of these (1,10,15,29,44). Most investigators agree upon the strong dependence of gas holdup on superficial gas velocity. Nevertheless, it is usual to find contradictions in many of the investigations and this is mainly because the authors have restricted themselves as far as equipment and systems utilized is concerned (1,40,44,50).

For example, Akita-Yoshida (1) neglected the effect of sparger diameter and column diameter (above 15 cm. I.D.) on gas holdup and also the liquid superficial velocity did not show any effect on holdup with values up to 4.5 cm/s. They have a correlation, with a linear dependency on the superficial gas velocity (NFr) as follows :

$$\frac{EG}{(1-EG)^4} = 0.20 \text{ NBo}^{1/8} \text{ NGa}^{1/2} \text{ NFr} \quad (11)$$

In the other hand Godbole et al (22) 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:

$$EG = 0.319 \text{ VG}^{0.476} \mu_L^{-0.058} \quad (12)$$

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 (15,18,29,32,38,43,44) agree with the conclusions from Akita-Yoshida (1). Others sometimes partially agree with some of their conclusions (3,64,66). Most investigators (14,22,39,40,48,50,56,57) disagree with their finding. Kastanek et al (39) disagree with the common idea that knowing holdup data for at least 300mm. I.D. column diameter is enough for scaling up (54) to larger diameters. They established a correlation after testing diameters from 50 to 1000 mm using air and water given by

$$EG = \gamma [k / (k+D)] [VG^{4/5} / (2VG + 20)^{7/15}] \quad (13)$$

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

Basically what happens is most of the literature comparisons are made using inconsistent sets of variables.

Knowing that the liquid velocity effect on holdup is negligible, Kastanek et al (38) derived a theoretical equation which takes care of the most important variables :

$$EG = \frac{[(1/q)(\rho_G/\rho_L)]^{1/n+3} D^{1/n+3} (M_G/\rho_G)^{n/n+3} k^{1+n/n+3} VG^{3/n+3}}{2^{3/n+3} 4^{n/n+3} (2VG + 0.2)^{n+1/n+3}} \quad (14)$$

and the equation depends only on the system to be investigated. Where n, D , and k are parameters and are found from EG values for different column diameters. The equation was tested with systems like Na_2SO_3 -air, water-air, ethyl alcohol-air, etc. and gave very good agreement.

According to Eissa and Schugerl (14) the best correlation for gas holdup is the one achieved by Hughmark (32) using a wide range of liquid properties ($\mu = 0.9-152$ cp, $\sigma = 25-76$ dynes/cm)

$$\text{EG} \propto \text{VG} \left[(62.4 / \rho_L) (72 / \sigma) \right]^{1/3} \quad (15)$$

This equation compares well from Akita-Yoshida's (2) and Towel's (64) results.

Kumar et al (44) criticized investigators that found differences in holdup while bubble column diameter was varied. They did not investigate the effect in a broad variety of columns but dealt with columns ranging from 5 to 10 cm in diameter. With a simple sparger they found only 6.7% dispersion with their data when the following correlation was applied :

$$\text{EG} = 0.728 \text{ VG}^* - 0.485 (\text{VG})^{*2} + 0.0975 (\text{VG})^{*3} \quad (16)$$

where

$$VG^* = VG \left(\frac{\sigma \Delta \rho \, g}{\rho_L^2} \right)^{1/4}$$

Also using different spargers in a large diameter columns (16in.), Towell (64) found a really simple correlation for holdup:

$$EG = \frac{VG}{VSb + 2VG} \quad (17A)$$

and detected only slight differences when the sparger was changed. VSb in this equation is the velocity of a single bubble that is approximately the slip velocity at low gas holdup. This equation is similar to the one found by Calderbank (7) :

$$EG = \frac{VG}{26.5} \quad (17B)$$

which does not correlate any effect of geometric variables. In another attempt, taking into account a broad variety of small column diameters, Hikita et al (29) did not find any appreciable effect of this variable on the holdup and found a correlation :

$$EG = 0.672 (VG \mu_L / \sigma)^{0.578} (\mu_L g / \rho_c \sigma^3)^{-0.121} (\rho_c / \rho_L)^{0.062} (\mu_c / \mu_L)^{0.107} \quad (18)$$

that also takes care of the nature of the gas used. They checked the above correlation with an air-water system using many gases and solutions of electrolytes and non-electrolytes, and found good agreement. Moreover, neglecting the viscosity and density factors in (18), data can be predicted within 15% error.

Miller (48) found a relatively high effect in the holdup from the variation of gas distributors in a bubble column with a quite large diameter and some different liquids. He found correlations that compared with others' data gave relatively high values for holdup e.g.:

for a single nozzle :

$$K = \exp[-3.13 + 1.689 \ln Z - 0.279 (\ln Z)^2] \quad (19)$$

for perforated plates :

$$K = \exp[-3.06 + 1.624 \ln Z - 0.237 (\ln Z)^2] \quad (20)$$

$$EG = (1 - Y_L) K \quad (21)$$

where $Z = N_{Re}^{1/6} N_{Fr}^{1/8} Y_L^{-1/4}$ and Y_L is the liquid volumetric flow fraction.

Kato et al (40) did not separate the results using single spargers and perforated plates. They correlated their data assuming a EG standard at any given conditions

and plotted Pe (EG/EG^*) vs NFr . They found similar behaviour for a large variety of experimental data with air-water systems, using perforated plate and single nozzle gas distributors.

Yatish and Shah (57) gave a good explanation for the variation of the holdup with the column diameter supported observation that with larger columns random circulation of eddies exist while in smaller ones these eddies are absent. These results are in agreement with those of Ueyama (66) from radial gas holdup measurements. Also Freedman and Davidson (19) worked with different column diameters in the bubble flow pattern using air and water and found that the maldistribution of gas reduces gas holdup and causes recirculation of liquid. Maldistributions are more likely in large column diameters (38,58). Nevertheless, Nakoryakon et al (50) conclude that this maldistribution especially present in the center and near the wall will eventually vanish if the liquid velocity is constantly increased. This strongly disagrees with the results by Kato-Nishiwaki (40) and Shah et al (58). The latter authors also explain the detrimental effects on reaction rates because of the reduction in the effective concentration of the species which affects the overall driving force.

Some investigators evaluated the dispersion and backmixing effects in bubble columns. The injection of salt traces (14) can be detected with electrical probes or with the help of heat transfer and sensitive thermocouples (4,15). The frequent recommendation for avoiding the

pronounced backmixing is the use of large orifices in the gas distributor plate (19). Presumably large orifices give more uniformity in the cross sectional area (54). Also agitation tends to give uniformity and increase the holdup as found by Fair et al (15).

Perhaps the best interpretation of backmixing in bubble columns was made by Joshi and Sharma (34). They give an equation for the velocity profile in a real bubble column with liquid recirculation and with the help of the definition of vorticity of Lamb (45) :

$$V = \frac{2\omega_0}{k_2 AD} \left\{ \left[\frac{\pi k \cos(\pi Z^*)}{H r^*} \right]^2 + \frac{2}{Dr^*} \left(\frac{dR}{dr^*} \right) \sin(\pi Z^*) \right\}^{1/2} \quad (22)$$

Lately, the use of porous plate by investigators is gaining more attention (4,12,13,54,55). Aoyama (4) found the holdup increasing linearly with gas velocity up to a specific velocity where holdup starts decreasing sharply and this coincides with the transition from bubble to bubble-slug patterns where coalescence takes place. This agrees with the results of Deckwer et al (12) and Schumpe and Deckwer (55). They found a holdup of approximately 0.22 for this maximum, at a gas velocity of 5cm/s. They also found a slight effect of liquid height in contrast to results when spargers are used as gas distributors. It is important to remark that with dispersion experiments these investigators had scattered data near this maximum value for

holdup and minimum for dispersion (4). These observations represent the transition from homogeneous to heterogeneous regimes. This transition is also observed at lower gas velocities when larger column diameters were used (4). Except for teflon porous plates, the material used for construction of porous plates does not have an appreciable effect on holdup (42,47A). The unwettability of teflon seems to give larger bubbles resulting in a lower holdup (43).

When the dynamic gas disengagement method (DGD) (70) is used to measure holdup, experimentalists agree that only the large to medium bubbles are important (17,22,68). Furthermore, the holdup due to small bubbles (17) is not appreciably affected by the gas velocity but this holdup diminishes with increasing hole diameter in the distributor. The inverse situation is present when holdup for medium to big bubbles is measured; it increases with gas velocity and hole diameter does not affect it (17,68). The holdup from the DGD method can not be used for scale up because it does not account for construction parameters (17) except when strong liquid circulation is a common feature.

Many processes that use bubble columns have solid particles carried by the liquid (11,16,65). Experimentalists have tried to find the effects of these particles in the holdup. For example, Deckwer (10) did not find any influence of solids particles on holdup while working with a system paraffin- Al_2O_3 - N_2 at low gas velocities, a system which is similar to a real mixture in

coal liquefaction. His data is really dispersed in relation to Akita-yoshida correlation (1) without giving any explanation. But on the other hand, Ying and Givens (70) and Kara et al (36), also working with low gas velocities found the gas holdup diminishing when solid particles are added. They did not detect any variation at high velocities.

Very good agreement exists in investigations involving electrolyte solutions (1,13,29). 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. For this, Akita-Yoshida (1) suggested same correlation as in equation (11) but with 0.25 as a coefficient instead of 0.20 for non-electrolytes.

Recent studies are focusing on the viscous effects in the gas holdup (6,7,11,14,18,22,55,56) because of the nature of real liquids used in practice (6,65).

Eissa and Schugerl (14) worked with glycerol-water solutions using a medium diameter column and found a more uniform distribution of bubbles giving higher gas holdups for viscosities up to 3cp. This was explained by observing that drag forces are not large enough at low viscosities so coalescence is less likely to happen. They plotted holdup vs. liquid viscosity for different gas velocities. Also they found that gas holdup diminishes with increasing surface tension. This effect was more pronounced at high gas velocities.

Working with CMC solutions Schumpe and Deckwer (56) 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 (18) and Buchholz et al (6). The last two researchers also found holdup values lower for CMC solutions than for water at low gas velocities. Schumpe and Deckwer (56) recognized that gas holdup decreases with CMC concentration in the slug regime.

There is some agreement on the presence of a maximum holdup when CMC solutions are tested with porous plates as gas distributors (11,22,56). This maximum, similar to other investigations (12,55), corresponds to the transition from homogeneous to heterogeneous regimes 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 (18), the holdup increases as the hole diameter is reduced.

For CMC concentrations higher than 0.8%, using a porous plate, Schumpe and Deckwer (56) suggest the following equation to be used only in the bubble flow pattern :

$$EG = 9.08 \times 10^{-2} VG^{0.95} \quad (23)$$

and with a perforated plate :

$$EG = 2.58 \times 10^{-2} VG^{0.876} \quad (24)$$

and in the slug pattern where the holdup does not depend on the type of distributor (18) :

$$EG = 3.22 \times 10^{-2} VG^{0.674} \quad (25)$$

and for diameters smaller than 10 cm :

$$EG = 4.04 \times 10^{-2} VG^{0.627} \quad (26)$$

This agrees with earlier conclusions about the dependence of holdup on column diameter (14,48,57).

Obviously, in the slug flow regime the holdup does not depend on the viscosity of the liquid phase, so knowing this characteristic, Deckwer et al (11) correlate data in CMC solutions for superficial gas velocity above 2 cm/s :

$$EG = 0.0265 VG^{0.82} \quad (27)$$

where almost no difference is present using different

distributors.

Godbole et al (22) 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 :

$$EG = 0.225 VG^{0.532} (u_{App})^{-0.146} \quad (28)$$

With this equation they predict data with only a 5% error. Using results from Schumpe and Deckwer for different column diameters (56) they correlate the overall data with :

$$EG = 0.239 VG^{0.634} D^{-0.50} \quad (29)$$

But paradoxically equation (28) gave better results when large columns were investigated.

EXPERIMENTAL TECHNIQUES OF HOLDUP MEASUREMENTS

A broad variety of methods for holdup measurements exist (66,68) but maybe the most important and relatively new method is the dynamic gas disengagement (22) that is attracting attention lately. Some of the most common

techniques are reviewed below.

A- Dynamic gas disengagement :

This method consists of measurement of the decline of the aerated height with time after the gas flow to the column has suddenly stopped. The method assumes that the behaviour of big bubbles is similar to the steady state conditions (68). The technique was developed because two different types of behaviour were noted within the bubbles swarm (17). The gas leaving the dispersion rapidly can be considered as the instantaneous transport (68) and the gas leaving subsequently, represented by very small bubbles is entrained by local liquid circulation. The technique needs a high speed response. In order to measure the instantaneous variations (22) a film camera or resistivity probes (68) can be used.

B- Bed expansion :

Measurement of the height of the bed with and without gas flowing is widely used when batch systems are present. However, at high throughputs, the height before gas is shut off is difficult to measure because of fluctuations (15) in the top of the aerated bed.

C- Quick closing valves :

This is the same method as bed expansion but with the liquid also flowing so both gas and liquid valves have to be closed at the same time and for this it is necessary to use

magnetic or solenoid valves (14).

D- Pressure drop measurements :

The pressure drop in a bubble column is related to the holdup (69) as follows :

$$EG = (\Delta \rho / \rho_L) \Delta h / \Delta Z \quad (30)$$

where $\Delta h / \Delta Z$ is the pressure drop in a specific column length. Therefore, this method allows the investigator to calculate the holdup at many positions along the column and have a better estimate (29) of axial variation.

E- Electrical probes :

For this method, measurements are based in the differences in electrical conductivity between the two phase within the bubble column. It has been found to be accurate because the device can be moved both axially and radially and detect the local holdup instead of the overall holdup as in most of the methods (43). However, inaccuracies arise because of the effect of the probe on the localized pattern at the probe.

BUBBLE SIZE AND INTERFACIAL AREA

These two terms are closely related (65). The better the observation of bubble dynamics, the more accurate the interfacial area can be determined.

Bubbles swarm and shapes :

The behaviour of large number of bubbles crowded together is different from that of isolated bubbles (65). The wakes of bubbles disturb each other giving random motion through the continuous phase (27) and the zig-zagging motion of an isolated bubble is no longer present. The random motion of the bubbles is damped by the viscous behaviour of the liquid at low gas velocities but not at high gas velocities (60).

The bubble shape varies with the gas velocity (46) and some specific shapes can be defined :

Spherical :

Bubbles are closely approximated by spheres, in this case, interfacial tension or viscous forces are much more important than inertia forces (8). Some oblate spheroids are considered spherical particles if the minor to major axis ratio lies within 10% of unity (8).

Ellipsoidal :

These are oblate bubbles although most of the time they are not a steady state shape because of their motion.

Spherical Cap :

These bubbles look very similar to segments cut from spheres or oblate spheroids of low eccentricity. Some authors call them mushroomlike bubbles (25).

The bubble deformation is primarily increased with gas velocity and Grace et al (25) give a graphical correlation for those shapes utilizing the Reynolds (liquid properties and U , D_e of the bubble) and Eotvos numbers.

VARIABLES OF INTERFACIAL AREA

The effects of many variables on interfacial area in bubble columns have been studied by a large number of investigators (13,32,49,64). It can be concluded that interfacial area depends on : Gas flow, height of the bubbled liquid, physico-chemical properties of the system, column diameter, gas distributor geometry, parameters affecting the bubbles motion in the column, and the presence of particles or traces of surface active matter as well (38,65).

With the exception of a few correlations (37,44,56) most of the investigators tend to use the known relation for interfacial area (65)

$$a = 6 EG / d \quad (31)$$

which assumes bubbles are spherical.

Most experimenters deal with the measurement or

observation of bubbles shape and its relation to an averaged bubble diameter (2,18,49,66). Discussion involving bubble size should be understood as being inversely related to interfacial area.

Koide et al (42) found that the nature of the gas used in bubble columns does not give any significant effect on the behaviour of bubbles. This simplifies the scope of any investigation as far as bubble size is concerned.

Knowing that between bubble and bubble-slug patterns (63), the interfacial area tend to reach a maximum (31,56) some authors tend to characterize this as a transition. Specifically, Otake et al (52) found a ratio standard deviation/average bubble size near 0.15 in this region while far appart in the slug pattern the same ratio is approximately 0.4. Also Nakoryakon et al (50) 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.

The principal problem is how to approximate the bubbles shape to an average diameter for a hypothetical bubble. Many authors have presented good approximations (2,6,9,25,31,66) of bubble diameter. For example, Davies and Taylor (9) 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.

Treybal (65) 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 :

$$d = \frac{2.344 \times 10^{-2}}{(VL / 1 - EG)^{0.67}} \quad (33)$$

Houghton et al (31) 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

$$De = 1.13 a \quad (34)$$

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 (17,18) can be neglected. The bubbles that are not a sphere can be approximated by an oblate spheroid resulting in good

agreement with Houghton et al (31). They found the mean volume-surface diameters followed a geometrical distribution function and gave a good estimate of the volume-surface diameter :

$$\frac{dV_s}{d\phi} = 1.88 (V_o/\sqrt{g d \phi})^{1/3} \quad (35)$$

They developed a good correlation for a broad variety of liquids in agreement with most of authors (32,39).

$$\frac{dV_s}{D} = 26 NBo^{-0.50} (NGa NFr)^{-0.12} \quad (36)$$

They found the mean diameter dV_s decreased when column diameter was increased. With the help of the Akita-Yoshida (1) holdup relation, equation (11), they found a correlation for interfacial area when holdup is below 0.14 :

$$aD = 1/3 NBo^{0.5} NGa^{0.1} EG^{1.13} \quad (37)$$

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 (17) and Ueyama et al (66). For the former, the use of the gas dynamic disengagement method (22) led to the inclusion of two main bubble sizes, one relatively large and another very small, (approximately 0.1mm) (22); and the calculation of the Sauter mean diameter (2,44) with the help of the holdup for both fractions (22) :

$$dVs = \frac{f \sum nki dki^3 + \sum nGi dGi^3}{f \sum nki dki^2 + \sum nGi dGi^2} \quad (38)$$

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. They also suggested an equation for interfacial area :

$$a = \frac{6 (Ek + EG)}{(1 - Ek - EG) dVs} \quad (39)$$

Using liquid holdup. This improved the correlation of their data.

However, they also calculated the interfacial area for large and small bubbles separately making it easy to

determine the low contribution of small bubbles to the overall interfacial area. This is in agreement with the conclusions of Godbole et al (22). Godbole et al (22) suggested them not being totally neglected because the accuracy of scaling up would be improved if they are accounted for.

Franz et al (17) using glycerol solutions found many contradictions within their own data. The only interesting feature they found is that 15% more interfacial area results by evaluating the true surface of large bubbles as rotation ellipsoids.

Another interesting way to measure the average bubble diameter was developed by Ueyama et al (66) who used the resistivity probe method. They divided the cross sectional area into multiple identical cross sectional area portions and calculated the average Sauter diameter for the overall cross sectional area by the equation :

$$\langle d_{Vs} \rangle = \sum EGi \Delta Si / (\sum EGi \Delta Si / d_{Vs} i) \quad (40)$$

They also calculated the arithmetic average diameter and found this value 1.5 times greater than $\langle d_{Vs} \rangle$. They found the largest bubbles in the center of the column and the smallest in diameter near the wall and both always increasing in diameter with gas velocity.

They noted that $\langle d_{Vs} \rangle$ might be smaller than the true

dVs when considering mass transfer between liquid and gas due to the fact that small bubbles near the wall were taken into account and those might be only recirculating in the column with little contribution to overall mass transfer (68).

Buchholz et al (6) remark that their mean bubble diameter data gave a trimodal distribution with the first ellipsoidal shapes appearing at superficial gas velocities near 3cm/s. Increasing the gas velocity, the distribution becomes bimodal with large and small bubbles only. They also found that increasing the column height, the bubble diameter increases with the negative effect for the interfacial area. They took data at three different axial positions finding almost the same size and behaviour using a photographic method. This axial behaviour agrees with results from Towell et al (64) and Deckwer et al (12), using gas spargers. The former, actually did not find any variation with gas velocity and attributed those results to a balance in coalescence and break-up. These latter authors found mostly oblate spheroides bubbles in their data and preferred to correlate the mean diameter to the diameter of a sphere of equivalent surface area.

Using a simple sparger distributor Frontini and Williams (20) found the interfacial area given by linear relation with the superficial gas velocity using the Sauter diameter for their calculations. Kumar et al (44) found correlations for the interfacial area related to the sparger hole diameter and gas velocity in two different sparger

Reynolds number ranges. Also they did not find any linearity :

$$100 < \text{Reh} < 2100$$

$$a(\text{Reh})^{0.425} (\angle do^2 / \Delta \rho g)^{1/4} = 13650 \text{VG}^* - 9.094 (\text{VG}^*)^2 + 1.828 (\text{VG}^*)^3 \quad (41)$$

$$\text{and Reh} > 2100$$

$$a(\angle do^2 / \Delta \rho g)^{1/4} \text{Reh}^{0.4} = 0.0437 \text{VG}^* - 0.0291 (\text{VG}^*)^2 + 0.005 (\text{VG}^*)^3 \quad (42)$$

where VG^* was defined in equation (16).

Data was correlated with a maximum 17% error for some two phase systems. They also developed quite complicated equations for the calculation of Sauter diameter (17).

Schumpe and Deckwer (55) found a strong increase in the mean Sauter diameter with increasing gas velocity but with perforated plates a somewhat pronounced decrease was noticeable. At low gas velocities the perforated plates always give higher values for mean Sauter diameter as expected and found by same authors (56). The interesting feature is that when high gas velocities exist, the mean Sauter diameter tends to level out at 3mm independent of the sparger used consistent with Deckwer et al (12). These authors results agree with those of Shulman and Molstad (60) with water-CO₂ and water-H₂ systems.

Many authors (3,7,26,31,36,42,54,64,67,70) conclude

that the presence of even traces of surface active agents reduces coalescence of bubbles and increases their rise velocities. Koide et al (42) 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 :

$$db(g \rho / \sigma)^{1/3} = 0.64 (NFr / NWe^{1/2})^{0.1} \quad (43)$$

Calderbank (7) remarks that there is a minimum size and any further addition of surfactants can not affect it. Anderson and Quinn (3) found the same effect while using electrolytes and larger holdups (3,70). Deckwer et al (13) found the interfacial area for electrolytes five times larger than without them, but on the other hand, the mass transfer coefficient decreased with the addition of electrolytes in such a way that KLa for electrolytes is 1.5 KLa for non-electrolytes.

Anderson and Quinn (3) also found problems in repeating the experiments especially holdup, even under closely controlled situations. They assumed that there is a hysteresis effect in passing from one pattern to another. Moreover, the same hysteresis effect was not repeatable in consecutive experiments.

New investigations of viscous effects and bubble behaviour (22,36,38,49,56) have been made due to the increasing importance in industry. Houghton et al (31) found that bubbles become more spherical while viscosity is increased, so application of equation (31) is more accurate. Kara et al (36) however found dissimilar results, when viscous solutions were studied. They found bubble disintegration at low viscosity and increasing bubble size and coalescence at high viscosities. However Peebles and Garber (53) noted little effect of viscosity on the bubbles motion provided that $Re_b \geq 3.1 (g \mu^4 / \rho \sigma^3 g_c^3)^{-1/4}$.

Kastanek et al (38) presented a relatively good equation for calculating interfacial area in viscous solutions. Where glycerol aqueous solutions were the principal systems :

$$EG / a = de / 4 \quad (44)$$

de = equivalent diameter.

with 18% as the highest error.

All these results were found in viscous Newtonian solutions, but for non-Newtonian solutions the results are not so different. For example, Nakanoh and Yoshida (49) found the bubble size decreasing with increasing viscoelasticity. They predict the Sauter diameter with a

Newtonian equation proposed to

$$dVs = 26 \text{ NBo}^{-1/2} \text{ NGa}^{-0.12} \text{ NFr}^{-0.12} \quad (45)$$

With the help of results from Yagi-Yoshida (69) and the inclusion of the Deborah number they give a good graphical correlation for the interfacial area as a function of mean Sauter diameter.

They conclude that the interfacial area and mass transfer coefficients in viscoelastic fluids seem to be smaller than in inelastic fluids. This is due to the fact that in viscoelastic fluids large bubbles mingle with very fine bubbles whereas in inelastic liquids they are relatively uniform in size.

Godbole et al (22) did not find the bubble flow pattern above gas velocities of 2cm/s for any CMC solution. Only below CMC concentrations of 0.15% was the existence of two bubble sizes observed. When they plotted VG/EG vs. VG a common intercept right after 0.25% concentration appears; this means the same bubble rise velocity, which partially agrees with results from Calderbank (7), who found the rising velocity independent of the viscosity.

Franz et al (18) found that by increasing the CMC concentration, the Sauter diameter became larger and the bubble size distribution broader for the same perforated plate. However, the concentration effect is reduced if the

perforated plate hole diameter is increased. Obviously, the smallest hole diameter with the least concentrated solution has the largest interfacial area, but at high concentrations no differences were found with the variation of perforated plate, in agreement with Schumpe and Deckwer (56).

These latter authors remark that with increasing gas velocity, at all CMC concentrations, the interfacial area monotonically increases, but with increasing CMC concentration there is a drastic decrease in interfacial area. Using a sintered plate, a sharp maximum for interfacial area is observed at $VG = 0.7 \text{ cm/s}$, and if this amount is required in the slug pattern, the gas velocity has to be increased approximately tenfold. The mass transfer coefficient was found almost constant with a value of approximately 0.013 cm/s , which is in contradiction with Buchholz results (6).

Maybe the best results were presented in a graphical correlation, where plotting a vs. $(VG / (M_{App})^{0.51})$ a straight line was achieved for all distributors and CMC concentrations giving the following relation :

$$a = 4.65 \times 10^{-2} (VG / (M_{App})^{0.51}) \quad (46)$$

where (M_{App}) is calculated following Nishikawa et al (51) correlations (equations 7 and 8).

As a conclusion, Schumpe and Deckwer (56) remark that

the gas velocity and effective viscosity are the only important parameters to be considered for interfacial area and holdup, provided that the bubble column diameter is large enough (11).

EXPERIMENTAL TECHNIQUES FOR INTERFACIAL AREA DETERMINATION

Many techniques are used for determining bubble size but only the most important ones are described.

Photographic method :

This is the most simple method. The special equipment needed is only a high speed camera in order to freeze the bubbles in their motion. Usually pictures are taken at the wall and high enough in the column to avoid entrance effects (7).

Resistivity probe :

This is a relatively simple method of size measurement. Bubbles in their rising motion touch two vertical sequentially positioned tips. These tips are connected to an electrical circuit and when the dispersed phase (gas) touch them, the electric conduction is less (66). With the time spent by the bubble in touching both consecutive tips and the distance between them, the bubble velocity can be

calculated. With the time duration of a bubble signal recorded in one tip, the diameter of the bubble can be calculated using its velocity.

The method is likely to have error assigned in neglecting proper couples of small bubbles signals, so it is possible to measure larger diameters than the true mean diameter (66). On the other hand, a big bubble (bigger than the gap between the tips) is found smaller with this method.

"Chemical" method :

This method is based on specific well known two phase chemical reactions (55). For example, a gas A is absorbed on a liquid B and an irreversible pseudo n^{th} order reaction takes place. Measuring the absorption rate per unit volume (RAa) and knowing also the kinetic properties of the reaction, the interfacial area can be calculated from :

$$RAa = a CA^* \left(\frac{2 k D CA^{n-1}}{n + 1} \right)^{1/2} \quad (47)$$

where CA^* is the equilibrium liquid concentration.

Fiber optics probe :

This is a relatively recent method of measurement (21). Fiber optics is well known by its good light transmission and the method takes advantage of this property. The light

is conveyed to the objective and if the probe is in a gas of low refractive index, the light is totally internally reflected and returned through the guide but, if it is immersed in a liquid of high refractive index, the light is transmitted and dissipated. Investigators take advantage of the transition from one mode to the other which produces a step change in reflected light intensity which is helpful to detect the passage of a bubble boundary (21). This principle is the same as that used in boroscopes. Their use would be advantageous in bubble columns due to their easier installation and their movability. Thus investigation of bubbles dynamics can take place in any specific position in the column. The bubbles motion can be seen through, and photographs can be taken.

All these methods have their own restrictions. Schumpe and Deckwer (55) say that the photographic technique in general gives bigger values of interfacial area than the "Chemical" technique, and the difference increases with the gas flow rate and is even greater when CMC solutions are used. At very low gas velocities both methods seem to agree. They also state that the photographic method underestimates the bubble diameter especially when spherical caps are present and also because of the assumption of radial symmetry.

Many investigators found very good agreement between photographic and "Chemical" methods (44,46), photographic and resistivity probe methods (66) and photographic with scattering light methods (7).

Buchholz et al (6) attribute the differences between "Chemical" and photographic methods to the fact that usually only the superior or front portion of the bubble participates actively in the mass transfer. Linek (46) investigated both methods and concludes that the interfacial area in the "Chemical" method is dependent on the oxygen absorption rate. This is due to the shrinkage of bubbles as a result of absorption.

III. EXPERIMENTAL WORK

Experimental setup description :

Figure (4) depicts the schematic diagram of the experimental set up.

The bubble column used is made of plexiglass and its inside diameter is 33.3cm with a wall thickness of 0.64 cm. The total height of the column is 5.40m, consisting of four 128cm long sections flanged together and the gas distributor system which occupies 19cm of the column. A total of twelve pressure taps were in the column, the lowest being 12cm above the gas distributor. The distance between the bottom five taps is 31cm, for the next six taps the distance change to 60cm and the last one has only 35cm separation. Every tap has a hole of approximately 0.15cm. The tappings were all connected to a manometer board by means of flexible polypropylene tubing. The manometric board consists of a glass U tube manometer containing carbon tetrachloride (CCl_4) as the manometric fluid.

Air was supplied by an existing compressor through a filter, pressure regulator, and rotameters. A control valve was used to regulate the gas flow rate and a magnetic valve for quickly stopping the air flow.

Aqueous CMC solutions were pumped from a tank, 0.64m in diameter, through a magnetic valve, filter, rotameter and control valve. Air entered the column at the bottom,

FIGURE 4

BUBBLE COLUMN SCHEMATIC

through the gas distributor region consisting of a 10cm long cylindrical section packed with 1.4cm long and 1.2cm diameter ceramic raschig rings followed by a 28cm diameter distributor plate. The plate is a porous plate made with a mean pore diameter approximately of 70 μm . The distributor plate was fitted, so as to make sure air entered the column only through the plate and not through its sides.

Liquid entered right above the gas distributor. The two phase mixture flowed up the column and the liquid returned to the tank overflowing on top of the column and the air vented.

Two 1/4" holes were made in the column one at 62cm and the other 141cm above the gas distributor; these holes were used to set a borescope at different radial positions in the column. The borescope is an Olympus A080 and an Olympus OM2 Camera was adapted to the borescope in order to take pictures of bubbles at different radial and axial positions.

Experimental procedure :

All the experiments were conducted with no liquid flow. The experiment started by filling the column with a specific CMC solution. In this case solutions from 0.25 to 2.0% in weight, which were prepared using CMC from Sigma Chemical Co. and distilled deionized water with a maximum of 10 ppm as equivalent sodium chloride.

Then, the air control valve was opened and the air pressure regulator set to approximately 50 psig. The air

flow rate was adjusted to a desired value and the system was allowed to equilibrate for about 5 min. At this moment the air flow rate, air temperature and pressure at the distributor were noted. The borescope was set inside the column at 141 cm above the distributor and 2.5cm from the wall and approximately 18 pictures taken with the focus distance set 2mm from the tip of the borescope. Photographs were also taken with the boroscope located at 5cm, 7.5cm, 12.5cm and in the center line of the column. In earlier experiments, the borescope was switched to the lowest axial position to see the variation in the bubbles size. Photographs of a ruler with 0.5 mm divisions were taken at the same focal distance as that set in the borescope in order to have a pattern of comparison for the bubbles size.

The temperature of the liquid was also observed.

The height of the bubbled bed was taken and then the magnetic valve in the air line closed and the air disengagement (22) monitored at 5sec intervals after the valve was closed. Samples of the solution were drawn at the beginning and the end of the experiment in order to evaluate physical properties as viscosity, surface tension and density.

The density was measured with a hydrometer. The flow curve of the solution was determined with a viscometer Haake RV-12, and the surface tension with a Fisher autotensiomat.

For every solution, experiments with 8 different air flow rates were conducted. The same procedure was repeated for the different solutions investigated.

Experimental errors :

The fluctuation in the air rotameter readings were negligible. But for high velocities and/or high CMC concentrations there were fluctuations in the bubbled bed height giving a corresponding error in average gas holdup of 7%.

Calculations of desired parameters :

The total gas holdup is calculated using the following equation:

$$EG = \frac{H_b - H_l}{H_b} \quad (48)$$

Where H_l represents the height of the liquid bed and H_b is the height of the bubbled bed.

The bubble gas rising velocity is calculated by:

$$V_b = \frac{\Delta EG}{\Delta t} \quad (49)$$

Where the ratio $\Delta EG/\Delta t$ is found as the slope in figures 11 to 15.

The interfacial area for unimodal distribution is found with:

$$a = 6 \quad EG/dV_s \quad (31)$$

Where dVs represents the mean Sauter diameter.

For a bimodal distribution the equation changes (see appendix A) to

$$a = \frac{6 EG1}{dVs} + 6 \frac{EG2 V2}{db V1} \quad (50)$$

Where db represents the equivalent diameter of the large bubble and $V2/V1$ the ratio of gas bubbles rising velocities with subindex 2 for the large bubble.

IV. DISCUSSION OF RESULTS

Gas Holdup :

Holdup showed a maximum with superficial gas velocity for the porous plate distributor. This maximum is shifted to lower gas velocities as CMC concentration is increased. However, for high CMC concentrations this maximum was obtained at the superficial Gas velocity of 1 cm/s. Also, the maximum in holdup tends to vanish with increasing CMC concentration (see figure 38).

The holdup for different CMC concentrations at the same gas velocity is always lower for the higher polymer concentration in the bubble-slug pattern. See figures 34 to 38.

Gas disengagement :

From figures (11 to 15) for gas disengagement it is important to remark that the higher the gas velocity, the steeper the slope for disengagement (from 0 to 6 sec). This means that the gas leaves the bubble column faster and the average size of the bubbles present in the column is bigger at higher gas velocities. Also it is interesting to realize that a constant slope throughout the gas disengagement for a specific velocity and a specific CMC concentration, represents a uniform bubble size. The holdup was found to

be a maximum in this particular case as was expected.

Franz et al (18); have found higher values for gas holdup, even using perforated plates. Differences are especially at high gas velocities with about 40% variation in most of the cases.

Using a sintered plate with 0.2mm pore diameter, Deckwer et al (11) found a maximum in holdup at the same gas velocities as this study, i.e., $V_G = 1$ cm/s. The value of holdup is always lower than that found in my experiments. For example using a 1.5% CMC solution they found $EG = 0.04$ as the maximum while in this study the maximum is 0.12. Similar differences were found by comparing my results with results from Buchholz et al (6) using a Cr-Ni stainless steel porous plate with 17.5 μ m as the mean pore diameter. Both experiments (6,11) were carried out in 14cm diameter bubble columns and low superficial liquid velocities which does not affect the holdup greatly (56).

In general, gas holdup values obtained from my experiments are very close to those of Schumpe and Deckwer(56). Their results, results from Buchholz et al (6) and my experiments exhibit similar curves for holdup vs. superficial gas velocity as far as CMC concentration is concerned. See figure (5).

Bubble Size and Interfacial Area:

Figures 16 to 25 show the radial variation for bubble size at different gas velocities and CMC

concentrations. In general, the bubble size is smaller near the wall and has its peak approximately at $R/2$. For the same gas velocity the bubble size increases with CMC concentration as can be seen from figures 29 to 33. However, at high CMC concentrations, the data is scattered and no specific behavior can be interpreted. Nevertheless, for a 1.00% CMC solution, mean Sauter diameter values are very close to those from Franz et al (18).

Bubble size did not vary significantly at different axial positions in the lower portion of the column. Two positions one at 62cm and another at 141cm from the distributor where tested and approximately the same bubble size was found. However, no other higher axial position was investigated although it was evident that at high throughputs coalescence takes an important roll.

For low CMC concentrations the formation of large bubbles starts at relatively high superficial gas velocities (figure 26). However, with highly concentrated solutions big bubbles are evolved with gas velocities as low as 1cm/s, which represents the absence of a well defined homogeneous flow.

At low gas velocities, for all the CMC solutions excluding 2.0%, a maximum in specific interfacial area was found. This maximum is achieved at a superficial gas velocity of 1cm/s for intermediate concentrated solutions (figures 8,9) and at relatively higher gas velocities for the more diluted solutions (figures 6,7). This maximum diminishes with increasing CMC concentration and its

variation is directly related to gas holdup variation as expected. For the lowest maximum, 1.5% CMC solution, a fivefold gas flow is required to achieve the same interfacial area at higher throughputs (figure 9), which demonstrates the convenience of the homogeneous flow.

Interfacial area is calculated over the entire gas velocity range taking into account unimodal and bimodal distributions, figures 6 to 10 show both of them. In both cases at low gas velocities for diluted solutions the interfacial area is calculated only with homogeneous Sauter diameter because of the absence of big bubbles. Obviously, the specific interfacial area calculated using bimodal distribution is lower than the one calculated assuming unimodal distribution and the main reason is the use of a lower value for the equivalent diameter in the latter. At high superficial gas velocities interfacial area calculated using bimodal distribution is more reliable.

Comparing results for specific interfacial area with the correlation from Schumpe and Deckwer (56) agreement is found at low and high superficial gas velocities for only highly concentrated CMC solutions whereas, for diluted solutions the agreement is at high superficial gas velocities as can be seen from figures 6 to 10. The specific interfacial area calculated from this correlation is always lower than interfacial area from this study.

The main difference in specific interfacial area between this study and Schumpe and Deckwer (56) is at low superficial gas velocities and diluted solutions, conditions

in which interfacial area presents a maximum. This difference can be explained by the fact that these investigators correlated data found with different distributors and mostly perforated plates. Thus their generalized correlation is principally for perforated plates.

For a 1.5% CMC solution using sintered plate Schumpe and Deckwer (56) found approximately 0.64 cm^{-1} as the maximum for interfacial area using the chemical method (13), coinciding with a superficial gas velocity of 1 cm/s . For the same conditions, the interfacial area is 1.4 cm^{-1} in this study. However, if gas holdup found by Schumpe and Deckwer (56) at those conditions and mean Sauter diameter found in this study are used, an interfacial area of 0.48 cm^{-1} results. Then, differences in gas holdup values are the main argument as far as interfacial area results is concerned. It is also important to remark that the difference in bubble column diameter (Schumpe-Deckwer 14 cm , this study 33 cm) could make any comparison wrong because of the coalescence effect in smaller diameters as well as the differences in the average pore diameter in the porous plate.

V. CONCLUSIONS AND RECOMMENDATIONS FOR FURTHER STUDIES

Bubble size and specific interfacial area studies were made in a 33cm diameter bubble column for the carboxymethyl cellulose (CMC) aqueous solution -air system using a 70 μ m porous plate gas distributor. The bubble column was operated in Bubble and Bubble-Slug flow patterns. The Bubble to Bubble-Slug transition occurred within superficial gas velocities of 1 to 5 cm/s depending upon the CMC concentration. The transition at higher superficial gas velocity occurred for the most diluted solution.

Gas holdup decreased with CMC concentration and exhibited a maximum at the Bubble to Bubble-Slug transition zone. The maximum diminishes with CMC concentration. Actually, this maximum appears to vanish for highly concentrated solutions. Results for gas holdup are in fair agreement with those from Schumpe and Deckwer (56) but having large differences for the maximum values.

Small bubble size was found to be smaller near the wall and bigger at $R/2$ in the column and it always increased with superficial gas velocity or CMC concentration. Finding bubble size distributions at high superficial gas velocities and/or highly concentrated CMC solutions was not successful due to some limitations in the photographic equipment in determining the larger bubble sizes. Therefore, results for specific interfacial area are not accurate under those conditions.

For low and moderate CMC concentrations it is recommended to work near the bubble to bubble-slug transition where the specific interfacial area exhibits a maximum. This maximum diminishes with CMC concentration and eventually vanishes at highly concentrated solutions.

The method for evaluation of specific interfacial area taking into account bimodal distribution presented in this study is more accurate than methods taking into consideration only a mean bubble diameter (6, 13, 17, 55) even if this average is calculated from accurate bubble size distributions as in Buchholz et al (6).

RECOMMENDATIONS FOR FURTHER STUDIES

Bubble columns have many applications in two and three phase flow. The effect of fluid properties and the nature of the gas distributor on flow pattern and interfacial area are important for the design and scale up of bubble column reactors. In the present work the effect of pseudoplastic properties in specific interfacial area using a porous plate was studied. Further work is recommended using other types of gas distributors and different methods for characterization of interfacial area in order to have a source of comparison. The use of a combination of borescope pictures and photographs from outside the column at high gas throughputs is also recommended. The objective of the

borescope can be enlarged with the use of a telex adaptor (from Olympus Co.), experiments using this device are also suggested.

VI APPENDICES

APPENDIX A :

Interfacial Area for bimodal distribution :

In any moment there are N_1 small bubbles and N_2 large bubbles present in the column for bimodal distribution. Large bubbles rise faster than small bubbles, then in any time interval Δt , N_1 small bubbles contacted the liquid in the column and $N_2 V_2 / V_1$ large bubbles did too so, the total interfacial area is defined by:

$$A_T = A_1 + A_2$$

Where A_1 and A_2 are the contributions for interfacial from small and large bubbles respectively.

Substituting A_1 and A_2 by their geometric definitions (per unit of total volume):

$$= \frac{N_1 \pi D_1^2}{V_T} + \frac{N_2 V_2 \pi D_2^2}{V_1 V_T}$$

In the other hand, gas holdup for each of the sizes becomes:

$$EG1 = \frac{\bar{N} D1^3 N1}{6 VT}$$

$$EG2 = \frac{\bar{N} D2^3 N2}{6 VT}$$

Substituting for VT in AT:

$$AT = \frac{6 EG1}{D1} + \frac{6 EG2}{D2} \frac{V2}{V1} \quad (50)$$

Where $V2/V1$ is found from figures 11 to 15 as the ratio of slopes for gas Disengagement between 0 to 5 sec and 5 to 60 sec, representing large and small bubbles respectively (22). $D2$ is determined from figure 26 which is a visual estimate of the average equivalent diameter of large bubbles. $D1$ is determined from evaluating photos using the mean Sauter diameter calculated with equation (38) only for small bubbles and is plotted in figures 29 to 33. Gas holdup $EG2$ can be taken from Gas Disengagement figures at 5 sec and gas holdup $EG1$ the difference between total holdup and $EG2$.

APPENDIX B :Experimental Data :

Table 1 to table 11

Figure 11 to figure 38

TABLE-1

Variation of holdup with time
for a 0.25% CMC solution

t,s VG,cm/s	6	11	16	21	60
1.0	0.014	0.024	0.035	0.049	0.063
2.5	0.025	0.051	0.081	0.119	0.149
4.1	0.047	0.094	0.141	0.193	0.218
5.6	0.071	0.114	0.157	0.205	0.228
6.6	0.048	0.077	0.116	0.164	0.194
7.7	0.077	0.106	0.140	0.178	0.198
8.6	0.076	0.105	0.144	0.173	0.195
10.1	0.074	0.098	0.157	0.186	0.206

TABLE 2

Variation of holdup with time
for a 0.50% CMC solution

t, s $VG, cm/s$	6	11	16	21	60
1.0	0.016	0.025	0.039	0.068	0.096
2.5	0.026	0.057	0.076	0.097	0.120
4.0	0.050	0.084	0.108	0.125	0.139
5.5	0.056	0.092	0.116	0.126	0.139
6.5	0.073	0.109	0.128	0.140	0.148
7.6	0.085	0.119	0.138	0.150	0.158
8.5	0.098	0.132	0.149	0.159	0.164
10.0	0.107	0.146	0.163	0.172	0.175

TABLE 3

Variation of holdup with time
for a 1.0% CMC solution

t, s $V_G, \text{cm/s}$	6	11	16	21	60
1.0	0.013	0.027	0.036	0.051	0.133
2.5	0.031	0.055	0.065	0.075	0.109
4.0	0.042	0.066	0.071	0.077	0.093
5.5	0.063	0.082	0.088	0.092	0.103
6.5	0.070	0.095	0.099	0.102	0.113
7.6	0.093	0.113	0.115	0.119	0.129
8.5	0.102	0.121	0.126	0.130	0.139
10.0	0.109	0.133	0.137	0.140	0.150

TABLE 4

Variation of holdup with time
for a 1.5% CMC solution

t, s $VG, cm/s$	6	11	16	21	60
1.0	0.017	0.027	0.036	0.046	0.120
2.5	0.031	0.041	0.043	0.046	0.059
4.0	0.046	0.058	0.060	0.062	0.072
5.4	0.065	0.075	0.077	0.079	0.086
6.5	0.073	0.085	0.088	0.090	0.098
7.6	0.090	0.100	0.102	0.105	0.111
8.5	0.093	0.107	0.109	0.110	0.117
10.0	0.103	0.117	0.119	0.120	0.127

TABLE 5Variation of holdup with timefor a 2.0% CMC solution

t, s $VG, cm/s$	6	11	16	21	60
1.0	0.017	0.018	0.018	0.018	0.023
2.5	0.031	0.037	0.038	0.038	0.045
4.0	0.049	0.054	0.054	0.054	0.056
5.4	0.062	0.066	0.066	0.066	0.067
6.4	0.069	0.073	0.073	0.073	0.074
7.4	0.077	0.083	0.084	0.084	0.086
8.4	0.087	0.092	0.093	0.093	0.095
9.8	0.098	0.101	0.101	0.102	0.103

TABLE 6

Shows the variation of Sauter bubble diameter
for different radial positions and gas velocities
for a 0.25% CMC solution

L/R VG, cm/s	0.15	0.30	0.46	0.76	1.00	D mm
1.0	2.0	2.4	2.3	2.6	2.2	2.3
2.5	2.6	3.0	3.4	3.3	3.1	3.1
4.1	2.1	2.6	3.2	3.3	3.1	2.9
5.6	3.5	3.3	2.5	3.0	3.4	3.1
6.6	2.4	3.6	4.5	4.2	3.0	3.8
7.7	2.4	3.0	4.4	3.6	5.4	4.0
8.6	2.6	5.1	3.2	3.9	3.9	3.9
10.1	4.9	4.0	3.4	2.6	2.0	3.9

TABLE 7

Shows the variation of Sauter bubble diameter
for different radial positions and gas velocities
for a 0.50% CMC solution

L/R VG, cm/s	0.15	0.30	0.46	0.76	1.00	D mm
1.0	3.1	2.2	2.8	2.1	2.7	2.7
2.5	3.3	3.7	3.6	3.2	3.3	3.5
4.0	3.5	3.1	2.7	2.5	3.7	3.2
5.5	2.1	4.1	3.7	3.6	3.0	3.4
6.5	3.2	4.9	4.5	4.1	3.9	4.2
7.6	3.2	3.1	3.5	3.3	2.6	3.2
8.5	3.5	4.3	4.8	5.8	4.4	4.7
10.0	4.3	7.0	2.9	4.9	3.0	5.4

TABLE 8

Shows the variation of Sauter bubble diameter
for different radial positions and gas velocities
for a 1.0% CMC solution

L/R VG, cm/s	0.15	0.30	0.46	0.76	1.00	D mm
1.0	3.2	4.5	4.1	3.9	4.0	4.0
2.5	4.7	3.2	4.4	5.0	4.4	4.4
4.0	4.4	4.1	3.8	2.9	2.1	3.7
5.5	1.4	2.5	6.8	2.9	1.7	4.2
6.5	1.3	2.5	2.8	4.3	4.6	3.7
7.6	1.3	2.4	4.2	1.7	3.2	2.9
8.5	4.4	2.4	3.6	1.5	1.4	3.5
10.0	3.5	4.8	2.7	4.5	2.5	3.9

TABLE 9

Shows the variation of Sauter bubble diameter
for different radial positions and gas velocities
for a 1.5% CMC solution

$\frac{L}{R}$ VG, cm/s	0.15	0.30	0.46	0.76	1.00	D mm
1.0	2.9	4.5	6.2	5.6	4.6	5.2
2.5	4.2	1.9	1.9	1.8	4.9	3.7
4.0	1.5	5.8	2.2	2.3	1.2	3.8
5.4	2.3	2.0	1.7	2.7	1.7	2.2
6.5	1.9	2.0	2.1	1.3	1.0	1.7
7.6	2.2	1.3	4.8	2.5	1.6	3.1
8.5	2.8	1.9	1.8	1.7	2.7	2.3
10.0	1.5	4.5	2.2	3.5	2.1	3.2

TABLE 10

Shows the variation of Sauter bubble diameter
for different radial positions and gas velocities
for a 2.0% CMC solution

L/R VG, cm/s	0.15	0.30	0.46	0.76	1.00	D mm
1.0	1.6	1.3	1.7	1.4	2.8	2.1
2.5	2.2	5.2	3.1	1.7	2.6	3.7
4.0	3.8	2.2	1.5	4.1	4.0	3.5
5.4	1.9	1.4	2.0	2.4	2.1	2.1
6.4	1.8	3.7	0.9	5.1	4.0	3.9
7.4	1.6	1.8	1.1	2.3	1.4	1.7
8.4	1.7	0.8	2.6	2.6	4.0	2.9
9.8	1.1	1.1	5.9	6.2	2.2	5.4

TABLE 11

Physical properties for different CMC
solutions at 22 C

Conc.	S.G.	Surface tension (dynes/cm)	Absorbance
0.25	1	71.5	----
0.50	1	71.0	0.15
1.00	1	72.0	0.27
1.50	1	72.3	0.41
2.00	1	72.3	0.52

FIGURE 11 :

GAS DISENCAEMENT FOR
A 0.25% CMC SOLUTION

FIGURE 13 :

GAS DISENCAGEMENT FOR

A 1.00% CMC SOLUTION

FIGURE 15 :

GAS DISENGAGEMENT FOR
A 2.00% CMC SOLUTION

FIGURE 19

BUBBLE SIZE RADIAL DISTRIBUTION
FOR A 0.50% CMC SOLUTION

FIGURE 21 :

BUBBLE SIZE RADIAL DISTRIBUTION

FOR A 1.00% CMC SOLUTION

FIGURE 22

BUBBLE SIZE RADIAL DISTRIBUTION
FOR A 1.50% EMC SOLUTION

FIGURE 23

BUBBLE SIZE RADIAL DISTRIBUTION
FOR A 1.50% CMC SOLUTION

FIGURE 25

BUBBLE SIZE RADIAL DISTRIBUTION

FOR A 2.00% CMC SOLUTION

SYMBOL V cm/s

◇ 6.5

† 7.6

x 8.5

Z 10.0

FIGURE 38

VARIATION OF HOLDUP FOR A
2.00% CMC SOLUTION

APPENDIX C :Maximum in interfacial area

A major contribution of this study is the indication of a maximum in interfacial area in the bubble pattern. Interfacial area is important in determining rates of heat and mass transfer.

The justification of the presence of this maximum is depicted in the sketches of figure 39. Sketch 39(a) indicates linearly increasing bubble diameters with superficial gas velocity. Gas holdup increases and reaches a maximum as superficial gas velocity is increased. See figure 39(b). The exception to this, as shown, is at high concentration of CMC. The solution becomes viscous enough to promote coalescence and retard break-up thus reducing gas holdup and eliminating the maximum. If we use the standard relation, that interfacial area is directly proportional to gas holdup and inversely related to bubble diameter, we obtain a maximum in interfacial area. See sketch 39(c). Thus, operation of bubble columns, with porous plates, in the bubble pattern, at low enough viscosities, should result in maximum interfacial area.

Figure 39 Sketch of maximum for interfacial area expected in this study.

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