STUDIES IN THREE PHASE GAS-LIQUID-SOLID FLUIDISED SYSTEMS

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The Study of Solid Behaviour in Three Phase
Gas-Liquid-Solid Fluidised Systems

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SUMMARY

The work is a logical continuation of research started at Aston some years ago when studies were conducted on fermentations in bubble columns. The present work highlights typical design and operating problems that could arise in such systems as waste water, chemical, biochemical and petroleum operations involving three-phase, gas-liquid-solid fluidisation; such systems are in increasing use. It is believed that this is one of few studies concerned with "true" three-phase, gas-liquid-solid fluidised systems, and that this work will contribute significantly to closing some of the gaps in knowledge in this area.

The research work was mainly experimentally based and involved studies of the hydrodynamic parameters, phase holdups (gas and solid), particle mixing and segregation, and phase flow dynamics (flow regime and circulation patterns). The studies have focused particularly on the solid behaviour and the influence of properties of solids present on the above parameters in three-phase, gas-liquid-solid fluidised systems containing single particle components and those containing binary and ternary mixtures of particles. All particles were near spherical in shape and two particle sizes and total concentration levels were used. Experiments were carried out in two- and three-dimensional bubble columns.

Quantitative results are presented in graphical form and are supported by qualitative results from visual studies which are also shown as schematic diagrams and in photographic form. Gas and solid holdup results are compared for air-water containing single, binary and ternary component particle mixtures. It should be noted that the criteria for selection of the materials used are very important if true three-phase fluidisation is to be achieved: this is very evident when comparing the results with those in the literature. The fluid flow and circulation patterns observed were assessed for validation of the generally accepted patterns, and the author believes that the present work provides more accurate insight into the modelling of liquid circulation in bubble columns. The characteristic bubbly flow at low gas velocity in a two-phase system is suppressed in the three-phase system. The degree of mixing within the system is found to be dependent on flow regime, liquid circulation and the ratio of solid phase physical properties.

KEY WORDS: Three-Phase Fluidisation, Gas Holdup, Solid Holdup, Mixing Index, Segregation, Flow Dynamics, Liquid Circulation Models, Solid Circulation Pattterns

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DEDICATION

This work is dedicated to:

.... the one that would have been most pleased, and greatly missed
-my mother, wherever you are.

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NOMENCLATURE

NOTATIONS	MEANING
A_{CS}	cross-sectional area of bubble column (cm^2)
C_f	feed concentration (g/cm^3)
C_T	total concentration (g/cm^3)
C_x	concentration at any point along column length
D_c	column diameter (cm)
D_s	axial solids dispersion coefficient (cm^2/s)
d_B	bubble size (μm or mm)
d_{Bmax}	maximum bubble size (mm)
dp	particle size (μm)
Fw	frictional force between fluids and wall
g	acceleration due to gravity (g/cm^2)
H_f	fluidised bed height (cm)
H_o	initial or unfluidised bed height
H/D	aspect ratio
L	column length (cm)
M	mixing index (v/v)
n	empirical constant
P	pressure of three phase bed system
P/VL	power input per unit volume of liquid (watt/cm3)
(dP/dz)	pressure gradient
R	dimensionless parameter ($\varepsilon g\ U_{SL}./(1-\varepsilon_g)U_{sg}$
U_{B0}	bubble terminal rise velocity (cm/s)
U_s	slip velocity (cm/s)
U_{sg}	superficial gas velocity (cm/s)
U_{sL}	superficial liquid velocity (cm/s)
u_s	bubble rise velocity (cm/s)
u_l	linear liquid velocity (cm/s)
u_s	linear convective velocity of solids (cm/s)
V_L	liquid volume (cm^3)
V_p	solid settling velocity (cm/s)
w, w_s	weight of solids particles
z/L	normalised column length

ε_g, ε_l, ε_s	gas, liquid, solid holdups
μ	viscocity (g/cm^3)
σ	surface tension (dynes/cm)
ρ	density (g/cm3)
Σ	summation sign

CHAPTER ONE

INTRODUCTION

1.1 PREVIEW OF WORK AT ASTON

Research into multi-phase fluidised system in bubble columns has been carried out at the University of Aston since the early 1970's. The initial group of reseachers, consisting of chemical engineers headed by Dr. E.L Smith and microbiologists supervised by Dr. R.N Greenshields, carried out work on bubble columns used for beer and alcohol fermentations, and biomass and metabolite production using moulds and bacteria. The engineering aspects (design, scaleup,and operation) for the bubble column fermenters for both aerobic and anaerobic processes were carried out by the chemical engineering group. Over the years, a number of studies has been carried out (Smith et al 1974, 1977, 1978, 1986), and the author's programme of research is a logical extension of this work.

1.2 SCOPE OF WORK

The purpose of this research work is described in terms of two main objectives, although both are inter-related. These are:

* to carry out a quantitative and qualitative investigation of the behaviour of bubble columns under "true" three-phase heterogeneous fluidisation conditions, with emphasis on phase holdups, mixing and segregation, and flow and circulation patterns, and,

* to integrate the knowledge of both two- and three-phase fluidised systems.

A comprehensive literature review of all parameters studied was carried out. The individual phase holdups in air-water and air-water-solids systems have been studied over a wide range of superficial gas velocities and for systems containing single, binary, and ternary particle mixtures. Both qualitative and quantitative analysis of the mixing and segregation were carried out. The influence of solids on these hydrodynamic parameters including bubble characteristics was examined using four types of solids (Ballotini, Styrocell, Diakon and Amberlite resin); of two sizes 600μm and 1200μm: particular attention was given to the effects of size, density, concentration, and "wettability". In the case of binary and ternary particle mixtures, the effect of the ratios of these variables was also investigated.

Experiments were performed in two- and three-dimensional bubble columns and standard techniques of measurement were adopted, with some modifications where necessary.

Conclusions have been drawn from a detailed analysis of the experimental data presented in graphical form, supported by visual observations and photographic evidence.

1.3 GENERAL INTRODUCTION

1.3.1 TWO - PHASE SYSTEM

A considerable amount of work has been reported in the literature covering investigations of various parameters affecting the design and operation of bubble columns using two-phase, gas-liquid (particularly air-water) fluidised systems. Also available in the literature are design data for complex two-phase systems to which solutes (e.g electrolyte) have been added.

The well studied air-water system was used in the author's work for preliminary tests and as a reference system for experimental data obtained from more complex three-phase systems.

1.3.2 THREE - PHASE SYSTEMS

The present work concentrated more on the three-phase gas- liquid-solid fluidised systems, using mono-sized particles of regular shape, which are easily fluidised. The reason for this is that many industrial processes consist of three or more phases. There are reports of research work in the literature concerning the hydrodynamic properties of such systems. However, the author has doubts about the fluidisability of particles used in some of these studies. Indeed, it was clear from the author's preliminary work that a proportion of the solids in certain systems may act as a "secondary distributor" for the dispersed gas because of the formation of a packed section of the solids at the bottom of the column. Moreover, a more definite pattern of the effects becomes apparent when higher concentration of the solids are present in the system: solids constituting up to 20% v/v of the total system were used in the present work.

1.3.3 COMPLEX THREE-PHASE SYSTEMS

Gas-liquid- solid systems containing two and three particle components were used for the experimental investigations. In some industrial processes, the systems contain more than a single solid and the solid particles may vary in size, shape and density. These properties often determine the fluidisability of the particle and could be expected to give rise, on occasion, to non-uniform mixing or segregation phenomena.

CHAPTER TWO

FUNDAMENTALS, CONCEPTS AND BACKGROUND THEORY OF THREE-PHASE HETEROGENOUS FLUIDISED BUBBLE COLUMNS

2.1 INTRODUCTION

Three-phase systems consist of gas, liquid, and solids which do not mix on a molecular scale. These systems are frequently used for processes where the aim is to contact two or more phases in order to carry out a chemical reaction or physical change.

The fundamental behaviour of three-phase heterogenous fluidised systems is primarily determined by the interaction between the phases. These interactions greatly affect the state of the dispersed phases, in general the gas phase and the solids, and make it difficult to predict parameters such as phase holdups and mixing. Furthermore, the occurence of agglomeration or coalescence of the dispersed phases and segregation in the case of solids, may not always be desirable from a processing point of view.

The selection of mathematical models and the formulation of empirical equations is another problem area. Related to this is the difficulty of evaluating parameters, such as phase holdups, required for these equations. The correlations used in the present work are discussed later in this chapter.

Visual observations constitute an important part of the studies of three-phase fluidised bubble columns. They provide insight into the fluid-dynamic phenomena associated with such systems, and aid in developing models .

2.2 PROCESS APPLICATIONS OF BUBBLE COLUMNS

Bubble columns are geometrically simple and provide a versatile way for conducting a variety of gas-liquid-solid operations with or without chemical reaction. They are also easy to operate and incur relatively low investment and maintenance costs.

Bubble column reactors provide process flexibility since they can be operated in both semi-batch and continous modes of operation, over a wide range of process temperature and pressure. Because there are no mechanically moving parts (e.g stirrers), and, hence, no sealing problems, they are especially suited for reactions involving aggressive gases as well as for high pressure and high temperature processes, e.g Fischer-Tropsch process: Saxena et al (1986), Stern et al (1985), Deckwer et al (1980, 1982). Additional advantages in such processes include: (a) a reduced rate of catalyst deactivation due to the washing effect of the liquid on the catalyst, and (b) high thermal capacity and reasonably constant bed temperature due to liquid circulation and liquid backmixing phenomena. Liquid backmixing is not studied in the present work; further information can be obtained from Krishna (1981), Baird and Rice (1975), Shah et al (1978).

In many bio-technological processes, floculated cells play an important role. The suspension of the cells is usually achieved in a three-phase bubble column by aerating at relatively low gas velocities because of the fragile nature of some microbial cells; Smith et al (1986), Smith and Greenshield (1971, 1974), Schügerl et al (1977, 1978).

Bubble columns have been modified in many ways to suit particular applications. Loop reactions are exemplified by the ICI pressure cycle fermenter and its deep shaft modification (Hine 1978); a horizontally sparged bubble column has been used for air oxidations of organic and inorganic component where high gas component conversion is not required and low pressure is desirable (Joshi and Sharma 1976); and a sectionalised bubble column has been used for continuous, single-cell protein fermentation processes where low back mixing is required (Schügerl et al, 1978).

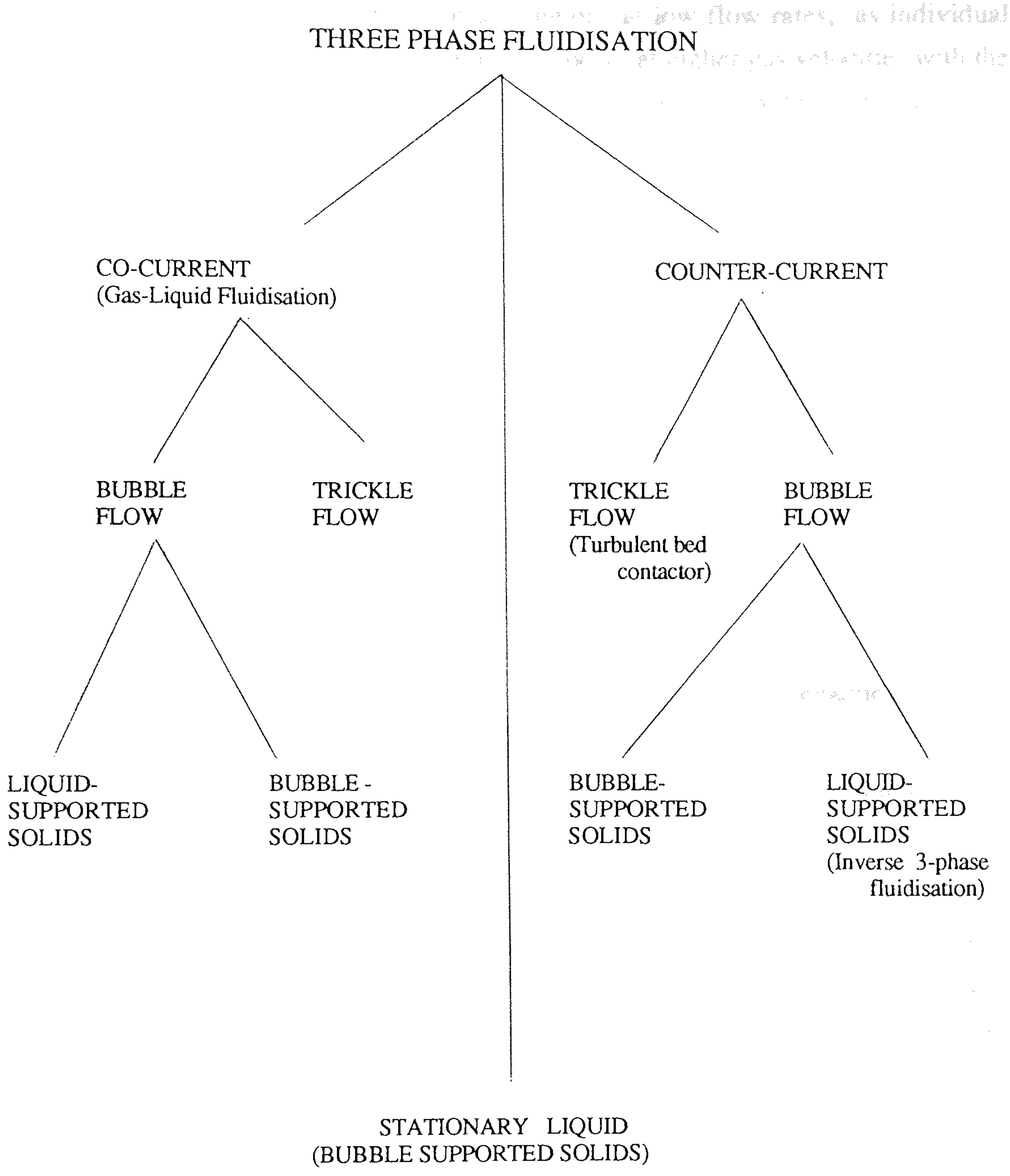
2.3 MODES OF FLOW

Figure 2.1 according to Epstein (1981), describes the fundamental distinctions between the various modes of operation of gas-liquid-solid fluidised beds, primarily in terms of the relative direction of the gas and liquid flows.

Modes of flow for three-phase fluidised beds have also been studied from the point of view of the behaviour of the gas phase. Alternative approaches for determining the operational mode have been based on studies of the axial concentration distribution of the particles in suspension bubble columns (Kojima 1981, Kojima et al 1981) and of axial variations of the pressure drop in solid-liquid fluidised beds into which bubbles have been injected. Therefore, the essential determinant of the mode of flow in a three phase fluidised bed appears to be the mutual interaction between the flow of the bubbles and the concentration and flow behaviour of the particles.

A comprehensive review of the various contacting mechanisms has been given by Muroyama and Fan (1985). Co-current upward flow of liquid and gas, with liquid as the continuous phase, was the mode used in the author's research.

Figure 2.1 : Taxonomy of Gas-Liquid-Solid Fluidised Beds (Epstein, 1981)



2.4 BUBBLE COLUMN FLUID DYNAMICS

When gas is dispersed into a liquid-solid bubble column to create a three-phase fluidised state, the gas will leave the distributor, at low flow rates, as individual bubbles of similar size and shape. Jetting will occur at higher gas velocities with the jets breaking up into bubbles of various sizes up the column. Bubble behaviour can often be used to characterise the fluid dynamic states in bubble columns.

2.4.1 FLOW REGIMES

Flow regimes are normally used to classify the fluid dynamic behaviour of gas-liquid and gas-liquid-solid fluidised beds, in conjuction with visual observations of the behaviour.

Three basic flow regimes exist, on the basis of the bubble flow behaviour, according to the classification of Wallis (1969) :

- dispersed bubbly flow , which is characterised by homogeneous distribution of almost identical, small bubbles

- churn-turbulent flow, where large bubbles move with high velocities among small bubbles, and

- slug flow, when large bubbles occupy the whole section of the column.

Figure 2.2 provides a schematic presentation of these flow regimes.

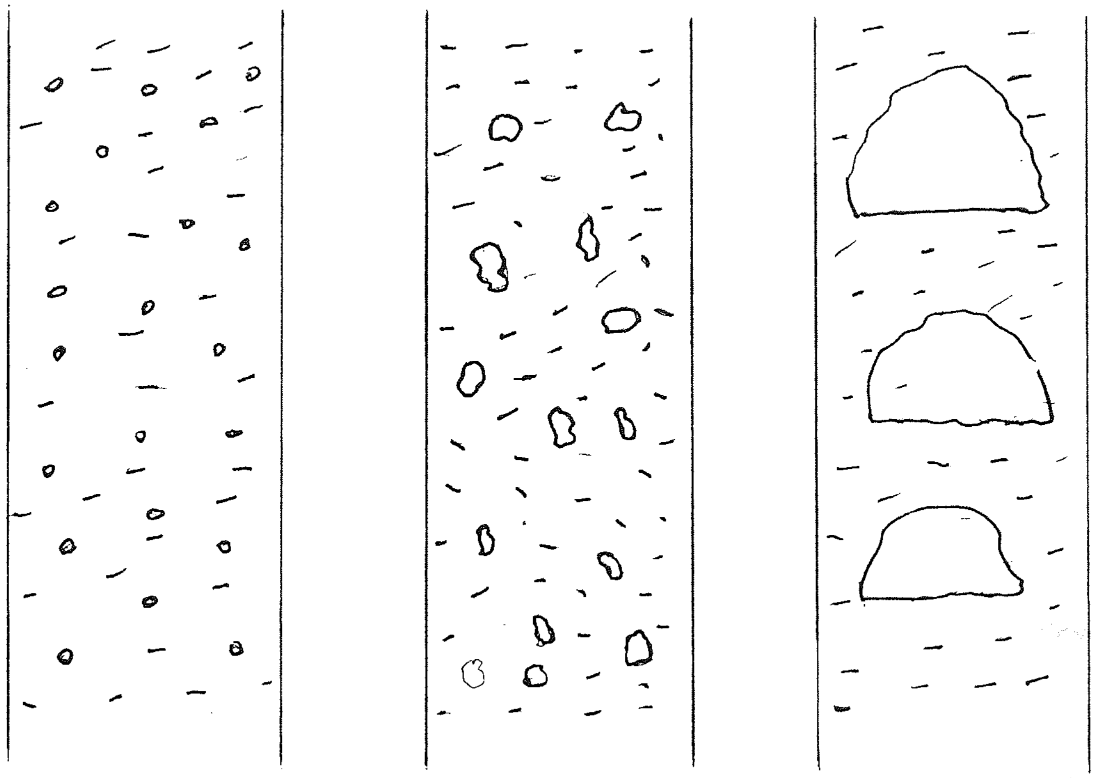
Govier and Aziz (1972) have indicated that at a gas holdup of 10% bubbles will only be about one bubble diameter apart. Therefore, bubble interactions even at a gas velocity as low as about 3cm/s can be expected and are observed. Bubble sizes will usually range from 2mm to 6mm in diameter, and shape will approximate to that of oblate ellipsoidal; the rise velocity of such bubbles is typically between 20 and 30 cm/s. At superficial gas velocities greater than 5cm/s, the formation of both groups of bubbles, bubble clusters and slug-like bubbles means that churn-turbulent flow eventually replaces the bubbly flow regime.

The following relationship is frequently used to account for the interaction between bubbles within the column

$$U_S = U_{B0} (1 - \varepsilon_g)^n \qquad 2.1$$

where, U_S is the slip velocity, given by equation 2.2 for cocurrent gas and liquid flow

Figure 2.2: Flow regimes in a bubble column (Wallis 1969)



BUBBLY FLOW | CHURN TURBULENT FLOW | SLUG FLOW

$$U_s = U_{sg} / \varepsilon_g - U_{sl} / (1 - \varepsilon_g) \qquad 2.2$$

and where, U_{B0} is the bubble terminal rise velocity which can be estimated from the graphical correlations of Grace (1973). Quoted values of the empirical constant, n, have varied from -1 to 2 [Govier and Aziz (1972), Lockett and Kirkpatrick (1975)]. For bubbly flow, $n = 1$ is often recommended and equation 2.3 is used for first design purposes:

$$U_s = U_{B0} \qquad 2.3$$

Smith et al (1986) indicated that a suitable value for U_{B0} is ~ 25cm/s for air-water systems.

2.4.2 BUBBLE BREAKUP AND COALESCENCE

At high superficial gas velocities, the nature and apparently chaotic changes in flow patterns in fluidised three phase bubble columns inevitably lead to bubble breakup and coalescence. The loss of interfacial area brought about by such coalecsence has an effect on mass transfer. Also, the coalescence-dependent transition between the bubbly and churn-turbulent flow regimes can be important in flow in pipes.

The size of bubbles leaving a sparger, d_B, has been found to depend on the design of the sparger used, operating conditions and physical properties of the liquid / slurry phase Smith et al (1986). For perforated plates, bubble sizes of the order of 4 - 6mm are common and can grow to several 'cms' in size depending on the operating conditions.

The maximum bubble size can be estimated by considering bubble breakup quantitatively using the relationship below :

$$d_{Bmax} = \sigma^{0.6} / (P / V_L)^{0.4} \rho_L^{0.2} \qquad 2.4$$

and the expression

$$P / V_L = \rho_L \, g \, U_{sg} \qquad 2.5$$

It has been suggested that if $d_B \neq d_{Bmax}$ bubble breakup or coalescence must be expected downstream from the sparger.

Recently, Chen and Fan (1989) proposed that a particle can penetrate a bubble when the value

$$d_p (\rho_s - \rho_L) g / \sigma_L (1 + d_p / 2R) > 6 \qquad 2.6$$

2.5 PHASE HOLDUPS

Evaluation of the axial holdups of gas, liquid, and solid phases in a three phase fluidised bed is usually performed by determining one axial phase holdup and obtaining the others by solving equations 2.7, and 2.8 simultaneously.

$$\varepsilon_g + \varepsilon_l + \varepsilon_s = 1 \qquad 2.7$$

and

$$-dp/dz = \{\{\rho_g\varepsilon_g + \rho_l\,\varepsilon_l + \rho_s\varepsilon_s\}g + \{\rho_g\varepsilon_g u_b\,(du_b/dz) + \rho_l\,\varepsilon_l u_l\,(du_l/dz) + \rho_s\varepsilon_s u_s(du_s/dz)\} - F_w \qquad 2.8$$

where ε_s, ε_l and ε_s are the axial phase holdups (i.e volume fractions) of gas, liquid,and solid respectively. The equations are based on one-dimensional steady state mass and momentum balances. At steady state, when the phase holdups are invariant with respect to axial position, the convective momentum term vanishes. The frictional loss due to fluid-wall interaction can also be taken to be negligible. Hence,

$$-dp / dz = \{\rho_g\varepsilon_g + \rho_l\,\varepsilon_l + \rho_s\varepsilon_s\}g \qquad 2.9$$

Direct measurements of gas and solid holdups are discussed in chapters 4 and 5. Extension of these relationships for systems containing binary and ternary particle mixtures are dealt with in chapters 6 and 7.

<u>THE CORRELATIONS OF EXPERIMENTAL DATA</u>

Correlations of experimental data for average gas holdups in bubble columns are numerous. However most are empirical in form and do not take account of the phenomena occuring in the column.

Gas holdup depends on the superficial gas velocity and can be sensitive to the physical properties of the liquid and solids present.

For air-water systems, employing perforated plate distributors a simple correlation

$$\varepsilon_g = 0.079\, U_{sg}^{\,0.4} \qquad 2.10$$

holds, with Usg in cm/s.

This correlation is applicable at high gas and liquid through puts for both 2-D and 3-D columns, the average error being ± 5%.

Fan et al (1985) and Shah et al (1982) have provided summaries of earlier correlations for the prediction of phase holdups in two- and three-phase systems. Four general correlations have been selected for comparison within the present work. These particular correlations were selected because they represent data covering wide ranges of values of system properties, and because two of them have been published relatively recently and make reference to the solids phase.

Begovich and Watson (1973)

$$\varepsilon_g = 1.15\, U_{sg}^{\,0.692}\, d_p^{\,0.107}\, D_c^{\,-0.037} \qquad 2.11$$

For air-water-glass beads (4.6 and 6.2mm), aluminium silicate (1.9mm) and plexiglas (6.3mm)

U_{sg}: 0-17.3cm/s, U_{sl}: 0 - 1.2cm/s and
Dc : 7.6cm and 15.2cm , and H_T : 2.2 - 4.5cm

Razumov et al (1973)

$$\varepsilon_s = 0.578 - 3.198\, U_{sg} - 0.538 U_{sl} \qquad 2.12$$

For the range U_{sl} = 1.25 - 5.0 cm/s and U_{sg} = 1.0 - 9.0 cm/s, using sand and slag beads (49 - 1270μm).

Reilly et al (1986)

$$\varepsilon_g = 0.296\, U_{sg}^{\,0.44}\, \rho_l^{\,-0.98}\, \sigma^{-0.16}\, \rho_g^{\,0.19} + 0.009 \qquad 2.13$$

For air-water-(varsol and trichloroethylene liquids) containing microbeads (420 - 1190 μm) at solids loadings up to 10% by volume.

Matsumoto et al (1989)

$$\varepsilon_g = U_{sg}\,(1 - R) / [\, 0.29\,(1 + 2.5\, \varepsilon_s^{\,0.85}) + 1.8\, U_{sg}\,(1 - R)\,] \qquad 2.14$$

Where R is a dimensionless parameter $[\varepsilon_g U_{sl} / (1 - \varepsilon_g) U_{sg}]$ and ε_s is the solids holdup, assumed to be constant throughout the column. The above equation holds for Usg: 1.0 - 30 cm/s, U_{sl} = 0 - 1.5 cm/s, ε_s = 0-30% and d_p = 66 - 1300μm.

It is the first equation to the author's knowledge to show the dependence of gas holdup on solids holdup.

2.6 LIQUID CIRCULATION PATTERNS

When a gas is sparged into a bubble column, the gas fraction is measurably highest at the centre, but relatively higher at the walls compared with liquid fraction particularly in the churn-turbulent regime. The radial density gradient arising from the variation in the gas holdup induces a buoyancy driven circulation, with upflow in the core and down flow in the outer annulus. This is a commonly encountered phenomenon and can therefore be said to be caused by the non-uniform holdup profile.

When there is ideal bubbly flow and no liquid feed, it is usually assumed that gross liquid circulation is absent. However, in reality, liquid mixing must take place as a result of the displacement of liquid by bubble movement and entrainment and exchange of liquid in bubble wakes. Where necessary, liquid circulation can be induced by non-uniform aeration, although usually at the cost of a reduction in the overall gas holdup.

The phenomenon of overall circulation in bubble columns is analogous to the occurence of convection motion in a fluid layer which is heated from below (Bernard cells). However, an important difference between the two phenomena is that there is a vertical density gradient in the case of a fluid heated from below : also the liquid properties will be changed at the same time. By contrast, liquid phase properties normally remain unchanged in the case of bubble columns.

Liquid circulation is considered to be primarily responsible for liquid mixing and has an important role in the distribution of solid particles in a three phase system. For systems containing two or more different types of particle, the liquid circulation has more complex effects, as will be discussed later in chapter 8.

Theoretical models have been put forward by several workers to describe circulation in bubble columns. Some researchers have accounted for liquid circulation by modelling overall holdup of gas in the column (Deckwer et al 1974, Shah et al 1982), but this approach is not general and cannot be used to indicate the actual fluid velocity in the column. Hills (1974) offered a model that used a force balance; Miyauchi (1981) suggested one that involves simplifying assumptions about the relationship between shear stress and velocity; and Clark (1987) developed a model for the distribution of axial velocity at half the height of the column. Models that take into account the visual observation of circulation in bubble column are listed on Table 2.1.

TABLE 2. 1: MODELS FOR LIQUID CIRCULATION IN BUBBLE COLUMNS

	MODEL	AUTHOR
1.	Laminar liquid circulation and bubble chain model	Crabtree &Bridgwater (1969)
2.	Laminar liquid circulation and bubble street model	Rietema & Ottengraph (1969, 1970)
3.	"Gulf - stream" model	Freedman & Davidson (1969)
4.	"Circulation flow model"	Miyauchi et al (1979)
5.	Liquid circulation model	Bhavraju et al (1978)
6.	Energy balance method	Whalley & Davidson (1974)
7.	Circulation cell model	Joshi and Sharma (1979)
8.	Recirculation model	Joshi (1980)

2.7 SOLID MIXING

Quantitative descriptions of the axial solids distribution in a three-phase fluidised bubble column have been based mainly on the axial dispersion -sedimentation model. This model takes account of a Fickian-type solids dispersion flux coupled with a solid flux due to the difference in the gravitational settling (sedimentation) and the convective slurry flux.

Many investigators have successfully used an equation of the form:

$$C_x / C_f = \text{Exp.} (-mLx) \qquad 2.15$$

for batch systems, where the model parameter, m, is defined as $(m = V_p / D_s)$; and $x = z / L$, the dimensionless column length.

The solid settling velocity, V_p, has been interpreted as either the particle terminal velocity or the hindered settling of a particle swarm relative to the liquid, the slurry or the column. Ds is the axial solids dispersion coefficient,which must be obtained experimentally; C_f is the feed concentration and C_x is the concentration at any point, x along the column length.

de Bruign et al (1989) recommended that the top concentration C_T must be used as a boundary condition rather than the feed concentration, C_f. If one takes the concentration at the top of the column as reference, then equation 2.15 becomes

$$C_x / C_T = \text{Exp.} [V_p (L-x) / D_s] \qquad 2.16$$

The model assumes no net flow of solids and constant gas holdup.

MIXING INDEX

Rowe et al (1972) introduced a mixing index, M, as a measure of the degree of solids mixing in a binary system. Accordingly, the mixing index is defined as:

$$M = \bar{w} / w \qquad 2.17$$

where $\bar{w}$, is the weight fraction of large particles in the upper section of the bed, and w is the weight fraction of the particles in the entire bed. M will vary from , 0, for the completely segregated state to 1, for the case of complete mixing.

The value of M will, of course, depend on the defined volume of the upper portion of the bed. In the present work, the upper section of the bed was defined as the volume occupied by the top 25% of the total bed working volume.

The concept of a mixing index was thus adopted on a volume / volume percent basis to indicate the degree of mixing of the particle components within the system, described in chapter 9.

CHAPTER THREE

SURVEY OF LITERATURE

3.1 INTRODUCTION

The literature on bubble columns is extensive, and experimental data on parameters such as phase holdups (particularly gas holdup), gas and liquid mass transfer, effective interfacial area, backmixing, heat transfer and power consumption have been reported. Earlier works in these areas includes that of Sharma and Mashelkar (1968), Deckwer et al (1974) and Baird and Rice (1975). Shah et al (1982), Muroyama and Fan (1986) and Olajuyigbe (1986) have provided reviews of the more relevant research, particularly that concerned with two phase, gas-liquid fluidised bubble columns. A selective review of literature relevant to the work is presented below.

3.2 REVIEW OF STUDIES OF PHASE HOLDUPS

The literature survey revealed that, despite the large number of theoretical and experimental studies made, important aspects of gas-liquid-solid fluidised bubble column have not been fully explored.

In the conventional study of individual phase holdups, overall values are often obtained through bed expansion measurements, and/or the pressure profile along the fluidised bubble column, the total amount of solid particles, and the continuity of the three phases,as exemplified in the publications of Akita and Yoshida (1973), Giopal and Sharma (1983), Kara et al (1982), Nottenkemper et al (1983), Jean and Fan (1986), Fan et al (1987), Olajuyibge (1986). These two techniques have been found to compare very well. Often, phase holdups are assumed to be uniform throughout the bed. However, information about local phase holdups can be, and have been, obtained by "in situ" measuring probes which have been used in many studies, in conjunction with pressure profiles (the manometric methods): see for example, Begovich and Watson (1978), Morooka et al (1982), Nottenkamper et al (1983), and Lee and de Lasa (1987). Nottenkamper et al (1983), in their measurements with an optical probe, obtained holdup measurement that were about 10% lower than those obtained from pressure measurements. However, it is inevitable that the hydrodynamics of a fluidised bed are somewhat disturbed by the use of probes despite attempts to minimise such effects. Generally, measurements using probes have indicated that there are variations in the phase holdups in both axial and radial directions in a three-phase fluidised bed. A gas disengagement technique has provided easier access to information on overall holdup structure especially for systems operated in the bubbly flow regime: Franz et al (1980); Vermeer and Krishna (1981), Godbole et al (1984) and Schumpe and Grund (1986), Patel et al (1987).

Olajuyigbe (1986) gave a comprehensive review of works concerning gas holdup characteristics in two-phase, gas - liquid fluidised bubble columns.

3.2.1 GAS HOLDUP

Earlier research includes that of Kato (1963), Ostergaard (1964,1965), Stewart and Davidson (1964), Adlington and Thompson (1965), Viswanathan et al (1965), Turner (1966) and Ostergaard and Theisen (1966). They all showed, as expected, that gas holdup increases with increasing gas velocity.

Various studies show that the gas holdup can be considerably lower in a gas-liquid-solid fluidised bed when compared with that in a solids-free gas-liquid system. Kato et al (1972), Ying et al (1980) and Vasalos et al (1980) investigated the effect of solids concentration and concluded that an increase in solids concentration generally decreases the gas holdup, although this effect becomes insignificant at high gas velocity (>10cm/s). Most work has involved relatively low concentration of solids, typically, less than 10% volume/volume concentration.

Quicker and Deckwer (1981) found that the decrease in gas holdup increased with the solids loading. Yang et al (1980) found that increasing the solids concentration from 15.7 to 45 wt % did not reduce the gas holdup any further. Kara et al (1982) reported that the gas holdup decreased with increasing solids concentration, but that the effect become less at higher solids concentrations and higher gas rates.

Kato (1963), in his studies of the effect of the solid particle size, density and amount of solids on gas holdup, reported a decrease in gas holdup with increasing particle size and amount of solids in the bed. Adlington and Thompson (1965) found that at higher gas velocities the presence of solids caused a decrease in gas holdups but this was independent of solid particle size.

Kelkar et al (1984) reported an effect of particle wettability on gas holdup: they found hydrophilic particles increased the bubble coalescence. Bhatia et al (1972) and Armstrong et al (1976), in their studies of the effect of solid wettability quantitatively described the role of wakes and bubbles in such systems. Kim et al (1975) indicated the existence of two distinct types of three-phase fluidisation: bubble-coalescencing and bubble- disintegrating fluidisation. The former occurs when the particles are smaller than some critical size, and the latter occurs when they are larger. Koide et al (1984) found the effect of solids on gas holdup to be larger in the transition region from bubbly to churn-turbulent flow than in the churn-turbulent flow region. Yasunishi et al (1986) reported that the presence of solids causes earlier coalescence of gas bubbles and postulated that this could account for the apparent absence of the zone of coalescence readily observed with two- phase systems.

A large number of correlations for gas holdup have been proposed in the literature particularly for two phase systems, see, for example, Muroyama and Fan (1985), Shah et al (1982). The various correlations show large scatter which has been attributed to the extreme sensitivity of holdup to the materials in the system: differences in easily measurable physical properties, such as density, viscosity and surface tension, do not provide sufficient explanation for the scatter. This problem prevents the use of a single universal correlation for gas hold-up.

Only a few correlations are based on a large number of experimental data. Begovich and Watson (1978) combined available gas hold-up points in three-phase systems and proposed an empirical correlation (See section 2.5). Their equation was restricted to batch liquid and solid systems. Ting et al (1980) applied the correlation proposed by Akita and Yoshida (1973) to their data and concluded that the correlation of Akita and Yoshida is equally adequate for three-phase systems.

The correlations used in the present studies, for comparison purposes, are more recent, and have been discussed in chapter two.

3.2.2 SOLID HOLDUP AND SOLIDS CONCENTRATION DISTRIBUTION

The axial variation of solids and solid holdup in a three-phase fluidised bubble column is often studied to provide information about solid distribution characteristics in systems containing single component particles, or solids mixing (or segregation) in those containing two or more particle mixtures. Earlier workers have included Cova (1966), Imafuku et al (1968), Farkas and Leblond (1969) and Kato et al (1972).

Kato et al (1972) measured the longitudinal concentration of solid particles and liquid mixing in 6.6, 12.2 and 21.4 cm diameter bubble columns using a diffusion model. The longitudinal dispersion coefficients of the solid particles and liquid based on the actual mean slurry velocity of solid particles and concentration of solid particles at the top of the column were correlated with dimensionless groups.

Kojima et al (1984) measured solids holdup in bubble columns with suspended solid particles. Air-water-glass beads in two columns of diameter 55 and 95cm were used. Their experimental results agreed well with the prediction made on the basis of the one-dimensional dispersion model.

Kelkar et al (1984) studied the hydrodynamics and mixing properties of gas, liquid, and solid flowing cocurrently upwards in a column. The effects of gas and slurry velocities, solids physical properties, particle size and solids concentration, interfacial tension, and slurry viscosity on phase holdups and axial dispersion coefficients were examined. The phase holdups as well as axial dispersion coefficients were not affected significantly by particle size, solids concentrations, and slurry velocities. However they suggested that wetting of the solids enhanced coalescence and decreased the axial dispersion coefficient when compared to the results for water.

Juma and Richardson (1983) successfully accounted for solids mixing for the case of liquid fluidised particle mixtures. Fan et al (1987) extended the idea of Juma and Richardson. Close examination of the solids concentration distributions in a three-phase fluidised bed containing two sizes of particles with similar density at superficial gas velocities > 0.9cm/s revealed the following distinct regions:

Region A : constant large and small particle concentration region, located at the top of the bed

Region B : constant large and small particle concentration region, located at the bottom of the beds, and

Region C : variable concentration region of large and small particles, located between region A and B

This classification is used in the present work to account for solids mixing within systems containing binary and ternary particle mixtures.

By far the most widely used representation of the solids concentration variation in the axial direction has been the "axial dispersion-sedimentation " model, first put forward by Cova (1966) and by Suganuma and Yamanishi (1966). The model requires two parameters - a solid axial dispersion coefficient and a particle settling velocity. Using experimentally measured axial concentration profiles, several workers have defined and obtained numerical values for these two parameters.

Smith et al (1986) proposed a modified dispersion-sedimentation model describing axial concentration distributions in a slurry bubble column. Their model has been used mainly to study the effect of polydispersed solids on axial solids concentration and particle size distribution. From their experimentally obtained axial solids concentration distributions for binary and ternary mixtures, they concluded that changes in gas velocity have only a small effect on the axial solids concentration distribution of completely suspended solid particles, and for polydispersed particles a greater variance in axial solids concentration distribution occurred even when having the same mean particle diameter as monodispersed particles. The axial particle size distribution for polydispersed particles indicated that significant segregation occurs for particle size ratios greater than two. These researchers also pointed out that a more uniform and larger specific interfacial area arises with monodispersed solids when compared with the case of polydispersed particles for a given mean particle diameter due to the effect of segregation.

A recent publication by Jean et al (1989) summarises various forms of dispersion-sedimentation model which have been used. These authors also concluded that the resulting numerical values of the model parameters are often not physically reasonable, especially for low slurry (liquid / solid) velocities.

Murray and Fan (1989) proposed a mechanistic model to explain solids dispersion in bubble columns. The model assumes solids dispersion can be explained by entrainment and detrainment processes in the wakes of rising bubbles. Their model leads to a functional relationship very similar to those obtained with the sedimentation-dispersion model.

Reilly et al (1990) obtained experimental measurements of axial solids concentration profiles over a wide range of solid and liquid flowrates normally encountered in bubble column reactors. They concluded that the axial dispersion-sedimentation model does not yield values of the two basic parameters of the model. They proposed a simplified model for the axial solids profile in three-phase bubble columns which requires only one experimental parameter. The model was justified through measurements of the settling velocities of particles in bubbly gas-liquid mixtures.

3.3 REVIEW OF STUDIES OF FLOW REGIMES IN BUBBLE COLUMNS

Flow regimes have been widely investigated, particularly in gas-liquid, two-phase fluidised bubble columns: Olajuyigbe (1986) has given a comprehensive review of literature in this area. The theory of bubbly flow (chapter 2) was developed by Lapidus and Elgin (1957), Richardson and Zaki (1954), and Wallis (1962). Their approaches give satisfactory correlations of gas-liquid flow only if bubbles are uniformly distributed over the cross-sectional area.

Characterisation of heterogeneous flow (i.e the churn-turbulent regime) has been studied by Hills and Darton (1976), Miller (1980), Shah (1982). They characterised the churn-turbulent regime as large bubbles moving with high rise velocity in the presence of small bubbles with the large bubbles taking the form of spherical caps with a mobile and flexible interface.

Kawagoe et al (1976) studied gas holdup as a function of flow regime and found a linear relationship with gas velocity in the bubbly regime, and that gas holdup was almost independent of gas velocity in the churn -turbulent regime. Bubbly flow may seem the most desirable regime since the bubbles are smaller and therefore interfacial area is comparatively greater (Guy et al 1986). However high gas flowrates often occur in industrial applications so that the churn- turbulent regime is reached (Bach and Pilhofer 1978).

The behaviour of a single bubble in a solid -liquid system was first investigated by Massimilla et al (1961). They measured the rise velocity of a single bubble of known volume injected into a 3.5cm ID cylindrical liquid fluidised bed of solid particles: 0.22mm silica sand, 1.09mm glass beads and 0.2mm iron sand were the solids used; water and air were employed as the fluids. They concluded that the relationship between the rise velocity and bubble size was essentially dependent upon the bed expansion and was relatively insensitive to the particle size.

In their experiments to characterise bubble swarms by measuring the size distribution of the bubbles emerging from the top of freely bubbly three-phase beds, they found that the mean bubble diameter increased with gas flowrate, the distance from the distributor, decreasing bed height. The latter parameter was found to be dominant, and they developed an empirical correlation which related bubble size to the distance above the distributor, the initial bubble size and the fractional expansion of the column. The qualitative results of Ostergaard (1966) support these findings.

Bubble coalescence and bubble breakup have been of particular interest as both occur simultaneously. Aldington and Thompson (1965) observed considerable bubble coalescence taking place in a three -phase fluidised bed containing different sizes of alumina particles. Lee (1965) reported that bubbles breakup in three-phase fluidised system when the size of the particles is comparable to that of the bubbles. Lee, Sharrard and Buckley (1974) postulated that bubbles split when they are penetrated by a solid particle. They claimed this occured when the Weber number , We, exceeded three (i.e $\rho_s U_B d_p / \sigma$).

Recently, Chen and Fan (1989) proposed a mechanistic model to predict the bubble breaking criteria between a single particle and a single bubble in a liquid medium. For bubble breakage the penetrating particle diameter should be larger than the height of the doughnut- shaped bubble. Also according to their criteria , a particle can penetrate a bubble when the value of $\{ (d_p (\rho_s - \rho_l) g / [\sigma_l (1 - d_p / 2R)] \}$ is greater than 6, where R is the radius of the bubble. Kim and Kim (1990) found that, in may practical systems, the value of this expression was about 26.

Kim et al (1975) reported the existence of a critical particle size which separates the bubbly flow regime and the churn-turbulent regime. The critical size for particles with density similar to that of glass was reported to be 2.5mm in diameter for air-water systems. Studies carried out by Muroyama et al (1981) and Fan et al (1982) indicate that besides the particle size, flow regimes also strongly depend on the liquid and gas velocities in the system.

3.4 REVIEW OF STUDIES OF FLOW CIRCULATION PATTERNS

Liquid circulation has been modelled as a one-dimensional process using momentum and a single phase "mixing length" by Ueyama and Miyaannchi (1979), Clark et al (1987), and Von Karman's hypothesis (1912) by Anderson and Rice (1989). Also, see Lockett and Kirkpartrick (1975), Shah et al (1982), Deckwer (1974). The applicability of these models can only be accurately assessed when the qualitative nature of circulation is better understood. Freedman and Davidson (1969) developed a liquid circulation model exhibiting a "gulf-stream" effect (also see chapter 8). They analysed their model mathematically by considering inviscid motion and sinusoidal distribution of vorticity. Field and Davidson (1980) improved further the vortex- pair model and proposed an explicit equation for the circulation velocity. Joshi and Sharma (1979) proposed multiple circulation cells in tall bubble columns, with cell height equal to
column diameter, without confirming their existence in the manner presented. More recently, Chen et al (1989) carried out a visual study of the effect of liquid depth on circulation in bubble columns. They observed a pattern of two rows of staggered circulation cells similar to the vortices shed behind circular cylinders studied by Von Karman (1912). The work of Chen and his co-workers shows similarities with that of the present study.

Devanathan et al (1990) have made the first attempt at tracing the liquid flow pattern in bubble columns by application of non-invasive, radioactive particle tracking (CARPT) . They monitored the motion of a single neutrally bouyant radioactive particle by an array of scintillation detectors and on-line computer to map the field. A more detailed discussion of some of the flow models that have been put forward by these researchers is presented in chapter 8 of the thesis.

3.5 REVIEW OF STUDIES OF PARTICLE MIXING AND SEGREGATION

Little work has been done on solids mixing in three-phase fluidised bubble columns. An experimental investigation to assess the rates of motion of the solid and liquid phases in a gas-liquid-solid fluidised system has been carried out by Evstropeva et al (1972); they measured average velocities and mean square values of the fluctuating velocity components for the rate of motion of light and heavy trace particles used to simulate the motion of the liquid and the fluidised particles. Criteria for the onset of stratification in the gas-liquid-liquid and gas-liquid-solid particle mixtures were defined experimentally by Epstein et al (1981). The incipient mixing (or segregation) process was viewed as a quasi-static balance of body forces.

A qualitative investigation based on visual observation of solid mixing in cocurrent three-phase fluidised beds containing a binary mixiture of glass beads was conducted by Fan et al (1984). They reported that the "complete " mixing state occurs largely in the coalesced bubble flow regime and slug regime and to a slight extent in the transition flow regime. The partial intermixing state occurs largely in the dispersed bubble flow regime and the transition flow regime, and slightly in the coalesced bubble flow regime. The complete segregation state occurs solely in the dispersed bubble flow regime.

Extensive effort has been made to model solids mixing in liquid-solid fluidised beds e.g Kennedy and Bretton (1966); Juma and Richardson (1983); and Patwardhan and Tein (1984). Fan et al (1987) modified the phenomenological model of Juma and Richardson to describe solids mixing behaviour and particle concentration distributions in the variable particle concentration region of a gas-liquid-solid fluidised bed (see section chapter 9). They characterised solids mixing by a mixing coefficient, and solids segregation, by a segregation velocity. Their approach was based on the empirical relationship developed by Jean and Fan (1986) between liquid velocity in the three phase system and the "equivalent" liquid velocity in the two phase, liquid-solid system.

CHAPTER FOUR

DESIGN OF EXPERIMENTS, EQUIPMENT AND EXPERIMENTATION

4.1 INTRODUCTION

To successfully achieve the objective of a research programme, careful design of the experiments, and consequently, the experimentation is a very important step. It is necessary to have a clear idea in advance, of exactly what is to be studied (i.e problem definition), what data are to be collected (i.e experimentation) and an understanding of how data will be analysed (i.e analysis).

Experimental design and experimentation for this work were carried out by considering the following:

(1) recognition and definition of the problem

(2) choice of factors (independent variables such as operating parameters)

(3) selection of the response (dependent variable - design paramaters)

(4) designing the experiments - materials selection, multi-phase systems, and suitable mode of operation.

(5) experimentation - including measurement techniques

(6) data analysis - results and their interpretation

(7) preparation of conclusions and recommendations

The final step, i.e the preparation of conclusion and recommendations, also involved the justification and comparison of results from analysed experimental data with relevant correlations and results obtained by previous researchers.

4.2 CHOICE OF MATERIALS

The choice of materials used in the experiments was based primarily on simulation of typical processes in which bubble columns are used in the biochemical, and chemical industries, the need to cover a range of physical properties, and constraints imposed by the hydrodynamic parameters investigated.

<u>SOLID PHASE</u>

Regular shaped, near spherical particle were used. The properties of the solids considered were:

. wettability - particle surface property, in terms of angle of contact with liquid.

. particle size uniformity, and

• particle density.

The solids used were

Ballotini and Styrocell	-	"non- wettable" solids
Diakon and Amberlite resin	-	"wettable" solids

For the experiments carried out in the two-dimensional bubble column, two particle sizes were investigated, 600μm and 1200μm, giving a particle size ratio of 1:1 or 1: 2 for the various particle mixtures. The exact particle sizes were obtained by using only those particles which blinded the required sieve size. This approach also ensured uniformity in the particle size. Non-uniformity in the particle size was simulated by mixing particles of different sizes in the desired ratio. Particle density variation was inevitable because of the choice of materials. It should be noted that the four chosen materials, especially ballotini, have been used in many previous experimental studies.

LIQUID PHASE

Tap water supplied from the mains was used as the liquid medium, which was also the continuous phase of the system.

GAS PHASE

Compressed air, available from the mains at 50psig, was used. The pressure was regulated before the gas entered the bubble columns as the dispersed phase.

4.3 CHOICE OF OPERATING PARAMETERS

The preliminary studies, mainly visual, of the behaviour of the bubble columns under varying superficial gas and liquid velocities led to the selection of the range of flow velocities best suited for the systems used. Also considered were the limits on the extent of bed expansion in the bubble columns and the pressure resistance of the material of construction of the bubble columns. The importance of flow regimes was also reflected in the choice of fluid velocities. The identification of different flow regimes is important to industrial applications and also helped in integrating knowledge obtained from visualisation studies in the present work with that from simpler systems previously studied by other workers.

4.3.1 SUPERFICIAL GAS VELOCITY

In biochemical processes, a high gas flowrate is not always desireable, Jamialahmadi (1982). The turbulent movement of bubbles or slugs may break up microbial flocs and have an adverse effect on shear-sensitive cells. Consequently, experiments must embrace the bubbly-flow regime. For the two fluids, i.e air and water, used in the present work, the literature survey revealed that departure from the bubbly-flow regime usually occurs at superficial gas velocities greater than about 4cm/s. The range of superficial gas velocity used in the present work was $0 \leq U_{sg} \leq 19.21$cm/s in the two-dimensional bubble column; and $0 \leq U_{sg} \leq 35$cm/s for the three-dimensional bubble column. These ranges of the gas flowrate covered the flow regimes believed to be of importance in industrial application. Previous Aston research workers, Shayegan Salek (1976), Jamialhamadi (1982) used relatively low values of superficial gas velocities, e.g $U_{sg} \leq 16$cm/s.

4.3.2 SUPERFICIAL LIQUID VELOCITY

Essentially, the systems were operated in the batch condition. Some studies were carried out to simulate a semi batch operation at superficial liquid velocity less than 1cm/s. The liquid flow rate is a very important parameter. It directly controls the output of the column and affects characteristics such as the concentration and residence time. Many biochemical reactions are relatively slow, and, in "once-through" processes, long residence time may be involved. High liquid velocities may lead to elutriation, and so create difficulties in maintaining high microbial concentrations inside the column.

4.3.3 TEMPERATURE

All experiments were performed at 24^{o}C with a variation of about $\pm 2^{o}$C. Measurements of the physical properties of all the materials used were also taken at room temperature. The measurements of particles density using the specific gravity bottle method (see section 4.6.1) were performed at a temperature of 20^{o}C using a water bath to control the temperature.

Jamialhamadi (1982), reported that liquid phase temperature has a significant effect on the gas holdup and bubble coalescence. However, very little has been presented about the subject in the literature. The effect of temperature was not investigated in the present work.

4.3.4 COLUMN DESIGN

All experiments were performed in two geometrically different column. The two-dimensional bubble column, figure 4.1, permitted detailed visualisation studies of systems behaviour and column hydrodynamics to be made. Such observations were essential to a clear understanding of complex phenomena such as mixing, segregation and phase circulation patterns. The two- dimensional bubble column also allowed a more accurate estimate to be made of the expanded bed height and hence of phase holdup. However, the marked wall effects associated with using two-dimensional columns mean that such studies must be complemented by work with three-dimensional systems, normally columns. Different sizes of three-dimensional bubble columns are commonly used. Details of the columns used by the author are given in section 4.4

4.3.5 PHASE PROPERTIES

Air and water were used as the fluids: their various properties are recorded in table 4.1. The physical properties of the solids are presented in table 4.2.

4.4 EXPERIMENTAL EQUIPMENT

All experiments were performed in the two- and/or three-dimensional bubble columns, which have rectangular and circular cross-sections respectively. Visual studies were mostly carried out in the two-dimensional bubble column.

4.4.1 THE TWO-DIMENSIONAL (2-D) BUBBLE COLUMN

Visualisation studies are an essential part of the present work. The two-dimensional bubble column was used because it enables clear visual and photographic observation to be made. The 2-D bubble column is of dimensions 1.3cm by 15.3cm by 143cm, constructed from perspex. The distributor section and the bed section were bolted together using flanges. A support screen was placed between the flanges inside a rubber gasket onto which was placed the distributor plate. This arrangement provided an appropriate free area and excellent gas distribution. The detailed design is as follows:

* a 0.2cm thick copper plate drilled with 0.1cm hole on a 0.6cm square pitch (figure 4.2)

* a 100 mesh wire gauze

TABLE 4.1: EXPERIMENTAL CONDITIONS FOR GAS-LIQUID-SOLID SYSTEMS

PARAMETER	RANGE
Liquid Phase	
Water	$\rho_L = 0.9917 g/cm^3$
	μ = 0.010 g/cms (=poise)
	σ = 72.6 10^3 dynes/cm
Gas Phase:	
Air	ρ_g = 0.0013 g/cm^3
	(Air mains pressure 50 psig)
Solid Phase	
Wettable solidsl:	
Diakon	dp = 600µm and 1200µm
"Amberlite Resin"	dp = 600µm and 1200µm
Non-Wettable solids:	
Ballotini	dp = 600µm and 1200µm
Styrocell	dp = 600µm and 1200µm
Column	
two-dimensional (2-D) bubble column	Width = 1.3 cm Length = 15.3cm Height = 143cm
Three-dimensional (3-D) bubble column	Diameter = 15.2 cm Height = Variable
Operating conditions	
Superficial Gas Velocity	$0 \leq Usg \leq 19.21 cm/s$ (2D column) $0 \leq Usg \leq 32 cm/s$ (3D column)
Superficial Liquid Velocity	$0 \leq Usg \leq 1 cm/s$
Temperature	Room temperature: $22^oC \leq T \leq 26^oC$

TABLE 4.2: PHYSICAL PROPERTIES OF SOLID PARTICLES

Solid Particle	Chemical composition	Particle size (μm)	Particle density (g/cm3)	Packed bed voidage, ε_p
Ballotini	glass	600	2.84	0.5
		1200	2.92	0.5
Styrocell	polystyrene	600	1.11	0.43
		1200	1.23	0.43
Diakon	perspex	600	1.28	0.5
		1200	1.11	0.5
Amberlite resin IRA-400CI	ion exchange resin	600	1.20	0.4
		1200	1.13	0.4

Figures 4.1 and 4.2 show schematic diagrams of the two- dimensional bubble column rig and the distributor plate arrangement at the bottom section respectively.

Water from the mains supply [1] was introduced into the column through valve A using rotameter R1 (rotameter m-series type 7s). The mains compressed air [2] was available at 50psig and was reduced, using a pressure regulator, to 5psig. Air was then passed into the column through rotameter R2 (type 10A). The rotameter was calibrated by installing a turbine flow meter in the air line. This was compared with estimates using the manufacturers' calibrated rotamenter flow conversion charts. Facilities were available for measuring pressure drop along the side of the column. Eight tapping points were located at the side of the column and these were connected to a common junction. Tappings could be isolated using clips such that only one was in use at any one time. Also, on the other side of the column were five sampling points protruding in to the column to avoid wall effect.

4.4.2 THE THREE-DIMENSIONAL (3-D) BUBBLE COLUMN

The three-dimensional bubble column is a vertical, cylindrical column of variable height and of 15.2cm internal diameter. The 3-D bubble column was constructed from standard lengths of QVF 0.152m bore pipe. The lowest section comprised an unequal 'T' piece with 3.8cm bore side arm for introducing liquid. The top section comprised a top gas outlet and a side outlet for the liquid- solid mixture.

Figures 4.3 and 4.4 show the layouts of the 3-D bubble column rig. Water was introduced into the column from the mains supply [1] by opening valve D: rotameter R3 (type 10s) was used for metering the flow. The gas supply was obtained from a compressed air service main [2], fed through a pressure regulator to the metering section via a control valve E. The pressure was measured by calibrated pressure gauge, P. The metering section consisted of two rotameters, R4 and R5 (types 14A and24A): Rotameter charts were used for conversion of the experimental readings. The gas distributor consisted of a circular plastic plate of the same diameter as that of the column and drilled with 61 holes of 0.75mm diameter on a 17.4mm triangular pitch .

4.5 EXPERIMENTAL PROGRAMME

When carrying out experimental studies on three-phase gas-liquid-solid fluidised systems, it is very important that all parameters which may have an effect should be considered carefully.

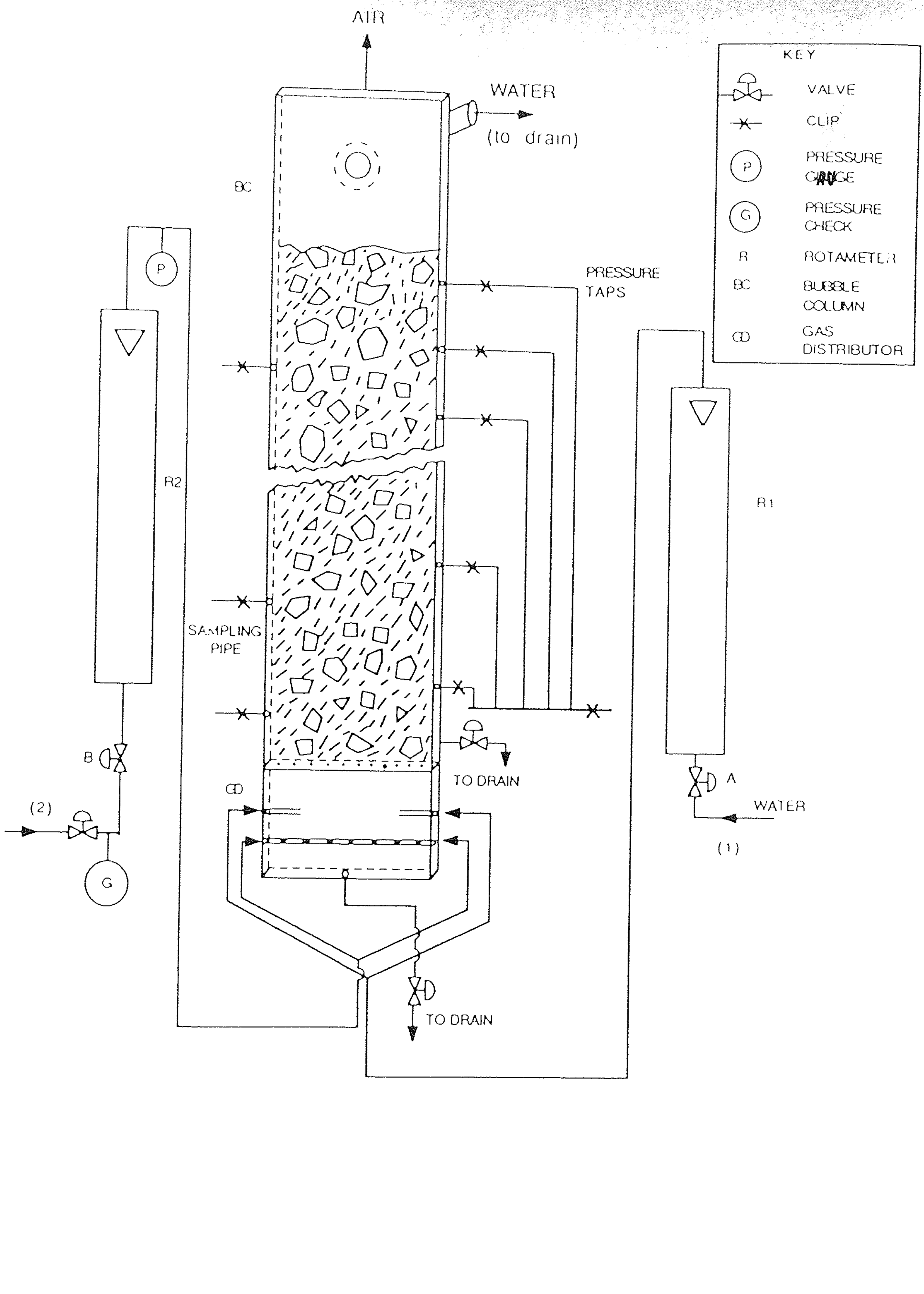
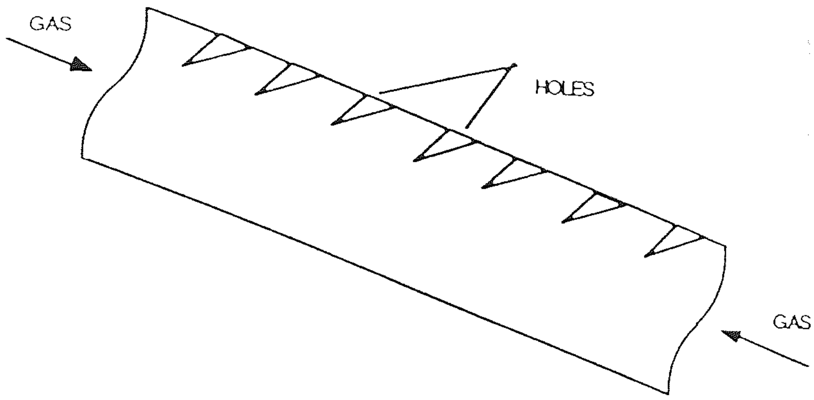
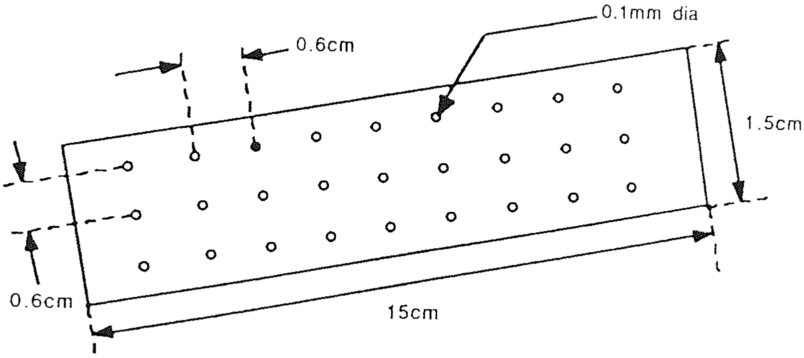


Figure 4.1: Flow diagram of the two-dimensional bubble column

(a)



INLET GAS TUBE



GAS DISTRIBUTOR PLATE

Figure 4.2 : Schematic diagram of the distributor design

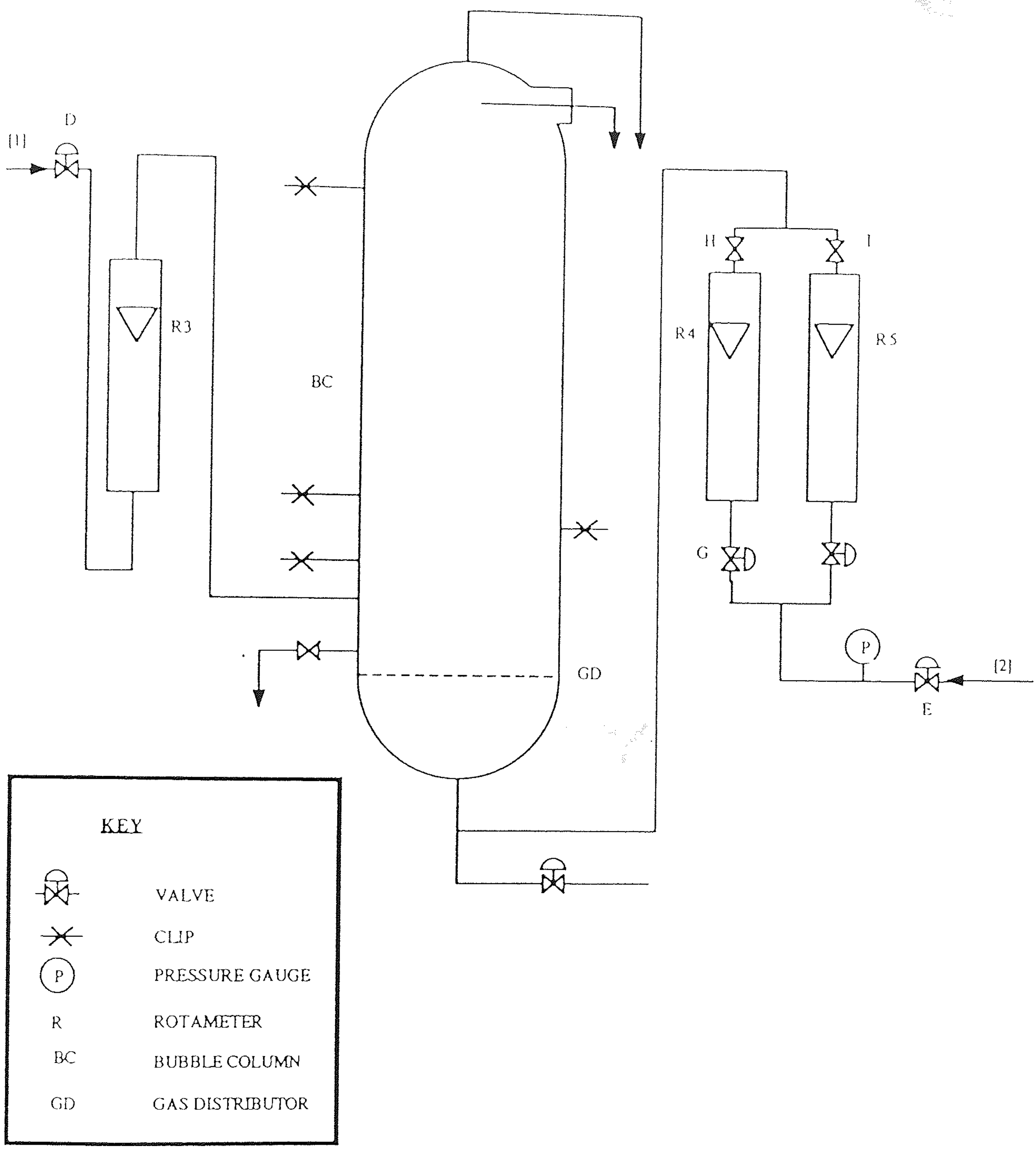


FIGURE 4.3: Flow diagram of the three-dimensional bubble column rig

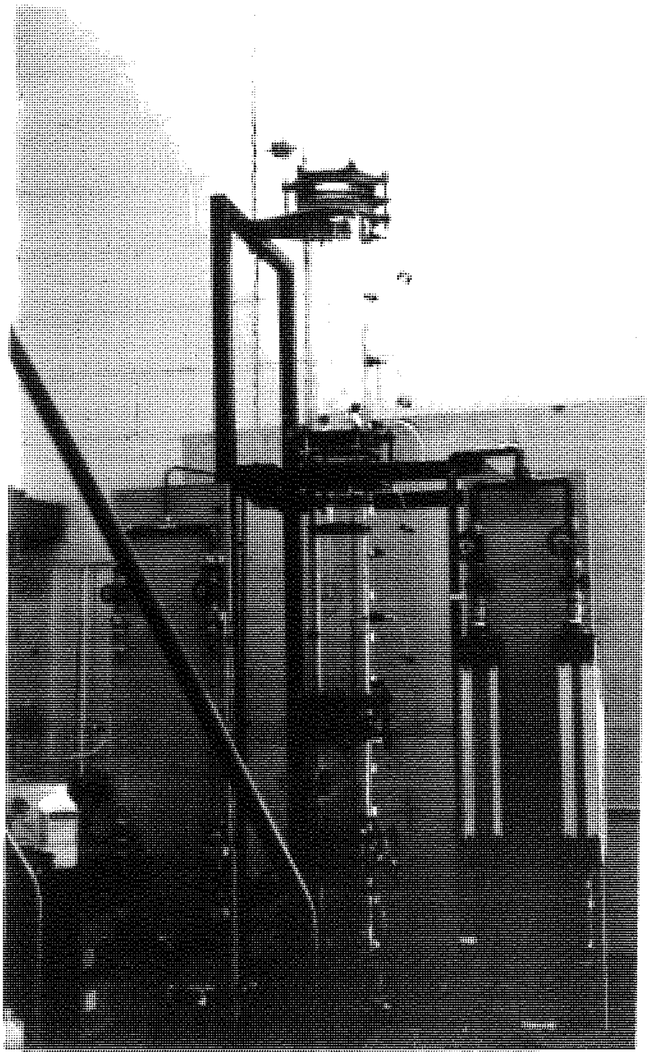


FIGURE 4.4: THE THREE- DIMENSIONAL BUBBLE COLUMN (PHOTO)

Because of the complexity of the study and to develop a systematic approach to experimentation, the experimental programme was divided into two parts:

(1) the quantitative experimental studies of the hydrodynamic design parameters, and

(2) the qualitative experimental studies of these parameters.

The latter involved visualisation studies, aided by the use of photographic and video techniques. However, these latter two are inter-related, and provided a clear and more accurate overall record of system behaviour.

The experimental programme commenced with work being carried out with the simple air-water system. These experiments were undertaken to investigate the effects of operating parameters, and system design on phase holdup (gas holdup only for the air-water system), flow regime and bubble dynamics.

The second stage of the experimental programme involved an extensive study of the phase holdups (gas and solid holdups), solid mixing phenomena, flow and circulation patterns in three-phase, gas-liquid-solid heterogeneous fluidised systems containing a single solid particle component. Again, the influence of operating parameters, system design, and physical properties of the solid phase was examined.

The third stage was concerned with the more complex three phase gas-liquid-solid heterogeneous fluidised systems containing binary and ternary particle mixtures. Additional operating parameters, such as solid mixture ratio, density ratio, and size ratio, and their effects on mixing and segregation phenomena were investigated.

Finally, visualisation studies were carried out on the systems investigated, as described later in chapter 8. Overlaps occurred with all the stages of the experiments, and this is reflected in the discussions.

4.6 EXPERIMENTAL MEASUREMENT TECHNIQUES

4.6.1 PHYSICAL PROPERTIES

Standard techniques of measurement were adopted as described below. All the methods used gave good reproducibility. The results reported are an average of three repeat experiments.

SOLID PARTICLE SIZE

A standard sieving method was employed. For precise particle sizing, only the particles that blinded the required sieve size were collected and used. For average sizes, used particularly in the three-dimensional bubble column to make up the required amount in some cases, the size range between two sieves was used.

SOLID PARTICLE DENSITY

The densities were determined by a displacement technique using a standard specific gravity method.

PARTICLE TERMINAL VELOCITY

This was determined by measurements of the time taken for free fall through a known height in a liquid medium (i.e water in this case) .

4.6.2 HYDRODYNAMIC PARAMETERS -MEASUREMENT TECHNIQUES

AERATED OR EXPANDED BED HEIGHT

Reliability in phase holdup values depends on the accurate estimation of the aerated or expanded bed height under the fluidisation conditions, especially at high superficial fluid velocity when the system is highly turbulent. Two methods were used to estimate this parameter.

The first method was by simple visual measurement of the expanded bed height following introduction of the gas, i.e the **bed expansion method**. For accuracy and consistency in the readings, three levels of the fluctuating bed position were recorded and the average was taken as the expanded bed height at that superficial gas velocity. A diagramatic illustration of the technique is given in figure 4.5.

The second technique was used mainly as a check. It was based on the measurement of the pressure gradient over the height of the column i.e the manometric method. Air was introduced into the column containing either liquid or a liquid-solid mixture at the desired superficial gas velocities. When steady state was reached (about 2 minutes was allowed for each measurement), the pressure profile up the entire height of the column was measured through the water manometer. The expanded bed height at the set superficial

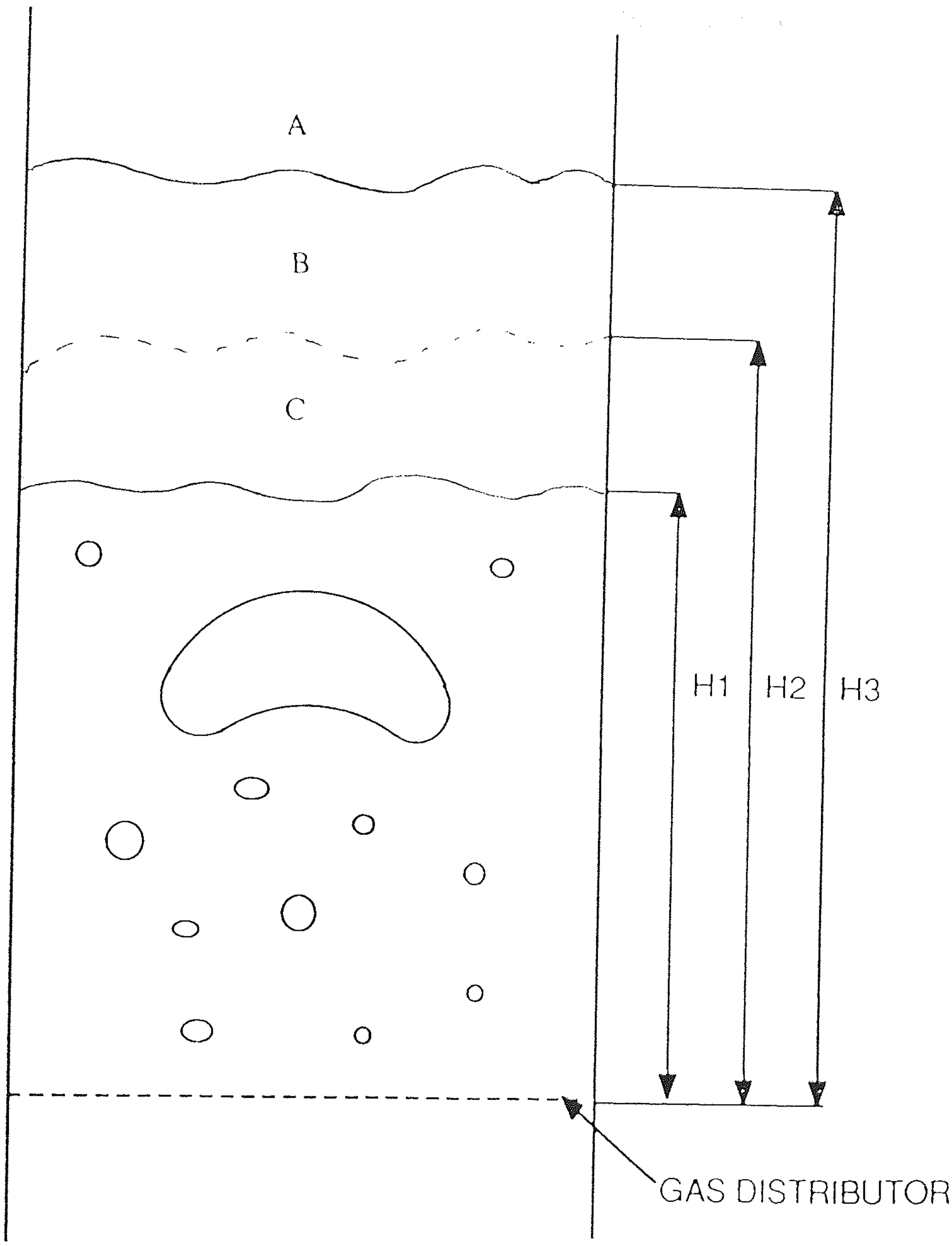


Figure 4.5: Diagrammatic illustration of aerated bed height, Hf; measured by bed expansion method

A -Expanded bed level at maximum fluctuation

B -Expanded bed level at intermediate fluctuation

C -Expanded bed level at minimum fluctuation

[$Hf_{average} = \Sigma\ Hf / n$]

gas velocity was taken as the point at which a change in the slope of the plot was observed, i.e the intersection point of the lines obtained from the pressure profile in the bed and the free-board region. Problems with fluctuation in the liquid level inside the manometer were encountered with this method. The fluctuations were dampened by pinching the tubing connecting the manometers to the column. Castros et al (1985) have used a photographic method to obtain simultaneous measurements of the levels of the undamped manoneters: their results for both damped and undamped systems agreed to within 5%.

SOLID CONCENTRATION

The concentrations were determined by withdrawing samples of the fluidised suspension into a 250cm^3 graduated standard bottle. Solid particles were then separated from the liquid and allowed to dry: the volume of the randomly packed dry particles was then determined. For multi-component particle mixtures, the individual particle components were separated and measured. The solids concentration was expressed in grams of the packed dry particles per cm^3 of the fluidised suspension.

GAS FLOW-RATE MEASUREMENTS

Superficial gas velocity was determined from the volumetric flowrate through the column, and the cross-sectional area of the column. Correction for the actual volumetric flowrate was accounted for by incorporating line pressure in the estimation: Appendix A provides the layout of the calculation procedure.

Moreover, the flow through a turbine gas flowmeter, installed in the compressed air main, was measured over a period of ten minutes at a fixed rotameter reading. The equivalent flowrate taken from the manufacturers conversion chart was also recorded. The two readings were compared, and the maximum deviation between the readings was approximately 3% .

CHAPTER FIVE

PHASE HOLDUP IN THREE-PHASE FLUIDISED SYSTEMS CONTAINING ONE PARTICLE COMPONENT

5.1 INTRODUCTION

Gas and solid holdups of gas-liquid-solid fluidised beds, containing particles of the same size and material, have been measured over a wide range of superficial gas velocities. Experiments were carried out in the two- and three-dimensional bubble columns, details of which have been presented in chapter 4.

THE SOLID PHASE

Eight sets of particles, comprising four different materials either 600μm or 1200μm in size were used: particle densities ranged from 1.11 to 2.92 g/cm^3. These sizes and densities were deliberate choices based on the author's awareness of the range of particles described in the literature, preliminary experimental studies and a review of process applications. The difficulty of fluidising large, and dense particles and their tendency to form a packed bed in the bottom section of the columns provided one practical limit. Interest in the use of bubble columns as fermenters in which the biologically active particles are typically from 500μm to 5000μm in size and have a density close to that of the liquid medium also influenced the choice of materials.

The properties of these solids and detailed experimental conditions have been discussed in section 4.3 and summaries are provided in tables 4.1 and 4.2.

5.2 EXPERIMENTAL PROCEDURE

A. OPERATING PARAMETERS

(i) The range of superficial gas velocity , Usg:

The values used were as follows:

Usg : 0 - 19.21cm/s [2D bubble column]

Usg : 0 - 35 cm/s [3D bubble column]

Pressure corrections were carried out on each measurement of superficial gas velocity taken from the rotameter tube, as discussed in chapter4, and outlined in appendix A.

(ii) The range of superficial liquid velocity, U_{sl}, used was <1cm/s.
Essentially, experiments were performed under batch conditions i.e
U_{sl} = 0cm/s.

B. SYSTEMS

The literature survey indicated the need to use a variety of solids. Ballotini has been used, and in a few papers reference is made to the use of polystyrene. Besides these two materials, particles of high density such as lead shot have been employed by some researchers.

The three-phase heterogeneous systems used in this part of the overall programme were:

* air - water - Ballotini
* air - water- Styrocell
* air - water - Diakon
* air - water - Amberlite

C. PHASE HOLDUPS

By employing bed expansion and manometric techniques, as discussed earlier in chapter 4 the bed height and, hence, the individual phase holdup were determined as follows:

From the bed expansion technique,

gas holdup ε_g, was obtained directly by the following equation:

$$\varepsilon_g = Hf - H_o / Hf \qquad 5.1$$

where, Ho and H_f are the bed height of the stagnant bed and fluidised bed respectively.

The solids holdup, ε_s was calculated using

$$\varepsilon_s = W_s / \rho_s A_{cs} Hf \qquad 5.2$$

where W_s is the weight of the particles in the system and ρ_s and A_{cs} are the particle density and cross-sectional area of the bubble column respectively, in consistent units.

The axial variation of the solids holdup in the bubble column was obtained by withdrawing samples along the length of the column. The dry concentration of solids in these samples were compared with the total dry concentration of the solid in the system at the start of the experiment. This method was also used to verify the results obtained using equation 5.2.

The sum of the individual phase holdups must equal to unity,

ie $\varepsilon_g + \varepsilon_l + \varepsilon_s = 1$ 5.3

Additionally, the experimental static pressure gradient is given by

$$-dP / dH = [\varepsilon_g \rho_g + \varepsilon_l \rho_l + \varepsilon_s \rho_s] g \qquad 5.4$$

and this provides a check on phase holdups, and was used in the case of gas holdup.

The holdup measurements were taken within two minutes of the start of the operation. This was sufficient to reach steady-state and to provide accurate and reproducible results by avoiding the accummulation of small bubbles, which may make a significant contribution to the gas holdup after lengthy aeration (Schugerl, 1981), especially at lower gas velocities.

5.3 EXPERIMENTAL RESULTS

The results from which the graphical relationships were obtained have been tabulated in appendix B1 - B8.

5.3.1 GAS HOLDUP

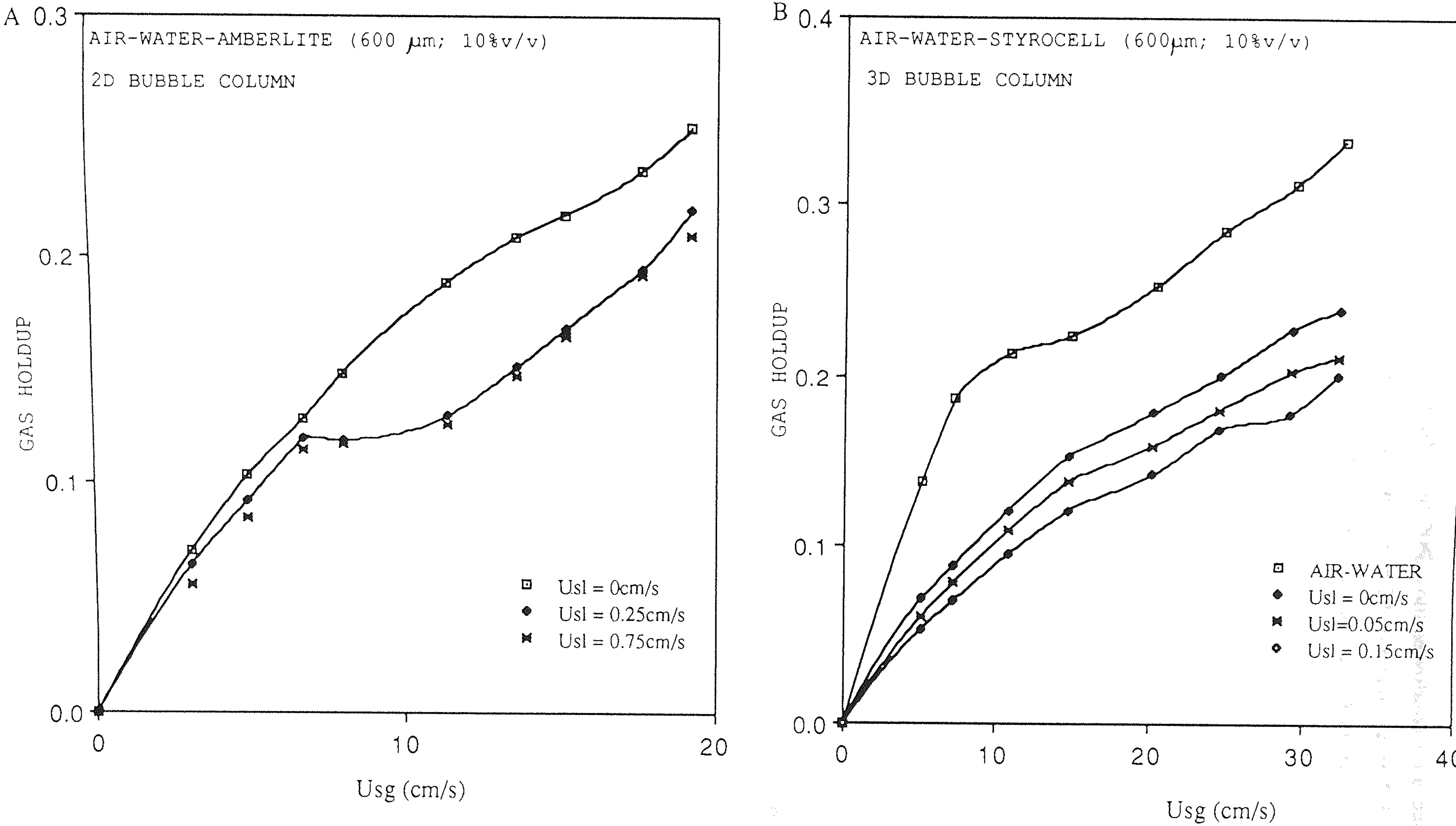
Generally, the gas holdup, ε_g, increased with increasing gas velocity, and the presence of solid particles reduced the gas holdup when compared with the solid-free system.

In the bubbly flow regime, typically for U_{sg} values < 5cm/s, a nearly linear relationship was obtained with the increase in gas holdup being relatively rapid. The gradient tended to level off at the higher superficial gas velocities.

For U_{sg} values >10cm/s, ie the coalescence and churn-turbulent regime, the characteristics of the gas holdup were found to be strongly dependent and more sensitive to several interacting factors; these are presented in the sub-sections that follow.

Simultaneous flow of liquid with gas in the columns caused a decrease in gas holdup. Also, as the liquid superficial velocity was increased a further slight decrease in gas holdup was realised. Figures 5.1A and B show results for the systems air-water-amberlite and air-water-Styrocell respectively, in both bubble columns.

Figure 5.1: Effect of gas and liquid superficial velocities on gas holdup



EFFECT OF SOLID CONCENTRATION

Results using solid concentrations of 10% and 20% volume/volume (i.e packed volume) of the total system are presented here. Figures 5.2A to F and Figure 5.3 show typical results obtained for systems containing Styrocell, Diakon and Amberlite, of two different sizes in the 2-D column, and for Ballotini in the 3-D column. Experimental data are listed in appendices B2 and B3 respectively.

Gas holdup decreased with increasing solids concentration. A strong dependence was noted up to concentrations of 15% v/v, the result for which often overlapped with those at 10% v/v. Above 15% v/v, the dependency tended to diminish and hold-up approached that for the air-water system at high values of the superficial gas velocity; this is particularly noticeable with 600μm particles. The exception was Ballotini; in this case, there was a further decrease at the 20% v/v concentration for both sizes. Figure 5.3 shows the result obtained for this system in a three-dimensional bubble column.

EFFECT OF SOLID SIZE

Using two chosen sizes, (600μm and 1200μm), the effect of solids sizes on phase holdup was investigated for all the solids. Figures 5.4 A to D illustrate typical results obtained by the direct measurement of gas holdup, detailed data being listed in appendix B, table B4

For air-water-Ballotini and air-water-Styrocell systems, a reducing effect on gas hold-up was produced with the effect more pronounced in the latter case. Similar results to those for Styrocell were obtained using Diakon and Amberlite: for details see table B2 in appendix B4.

EFFECT OF SOLID DENSITY

From Figure 5.5A to F, based on data in appendix B5, it can be deduced that increasing the solid density reduces the gas hold-up. The density ratio of Ballotini to Styrocell is approximately 2.4:1 and 2.6:1 for particles respectively 1200μm and 600μm in size. For Styrocell and Diakon (1200μm particles), the densities are very close at 1.23 and 1.11 g/cm respectively; the same is true for Amberlite and Diakon (600μm particles) at 1.20 and 1.28 g/cm^3.

Figure 5.2: Effect of solid concentration on gas holdup in the 2D column

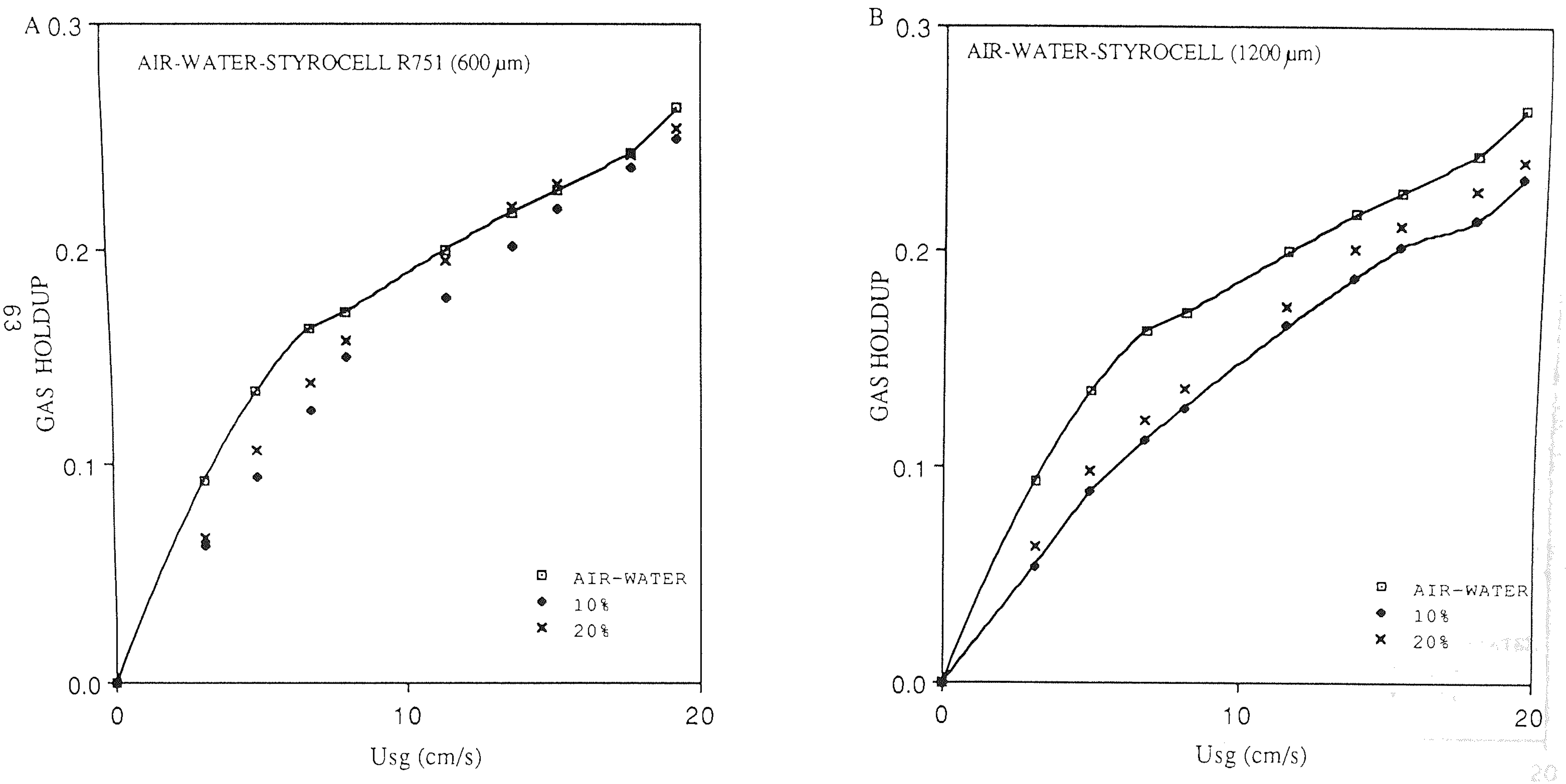
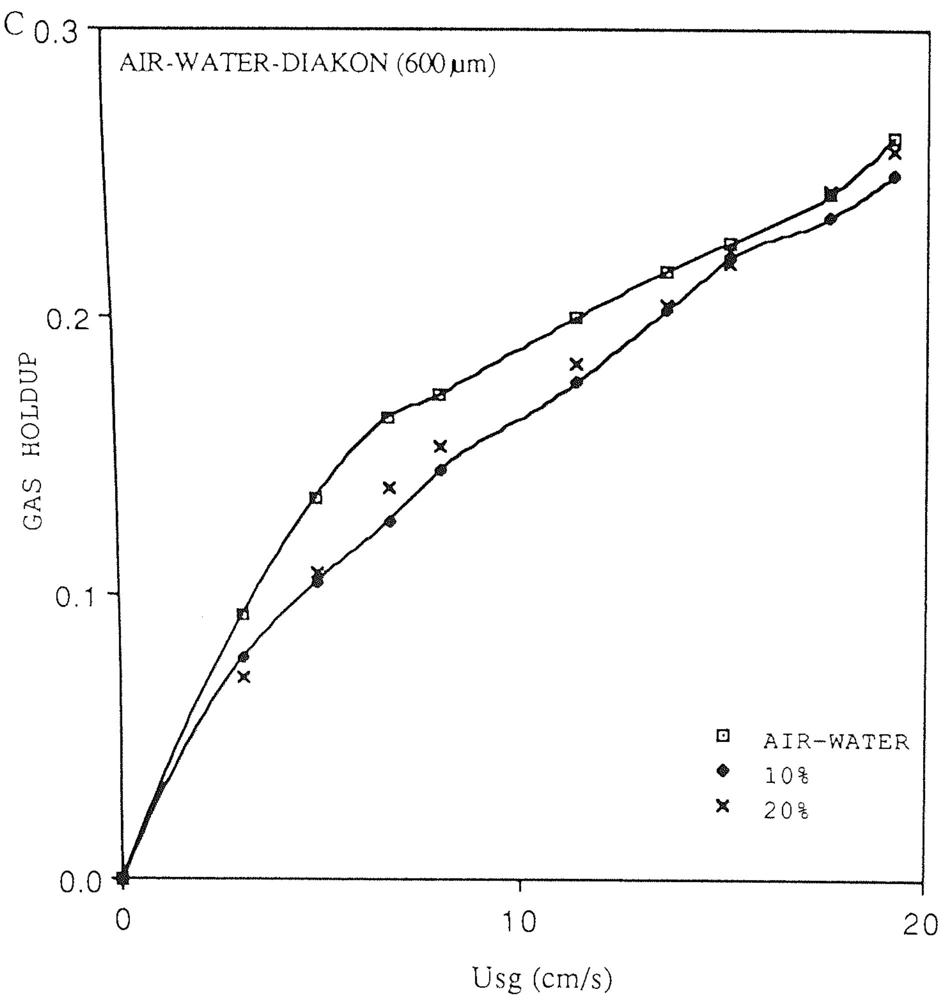


Figure 5.2 continued



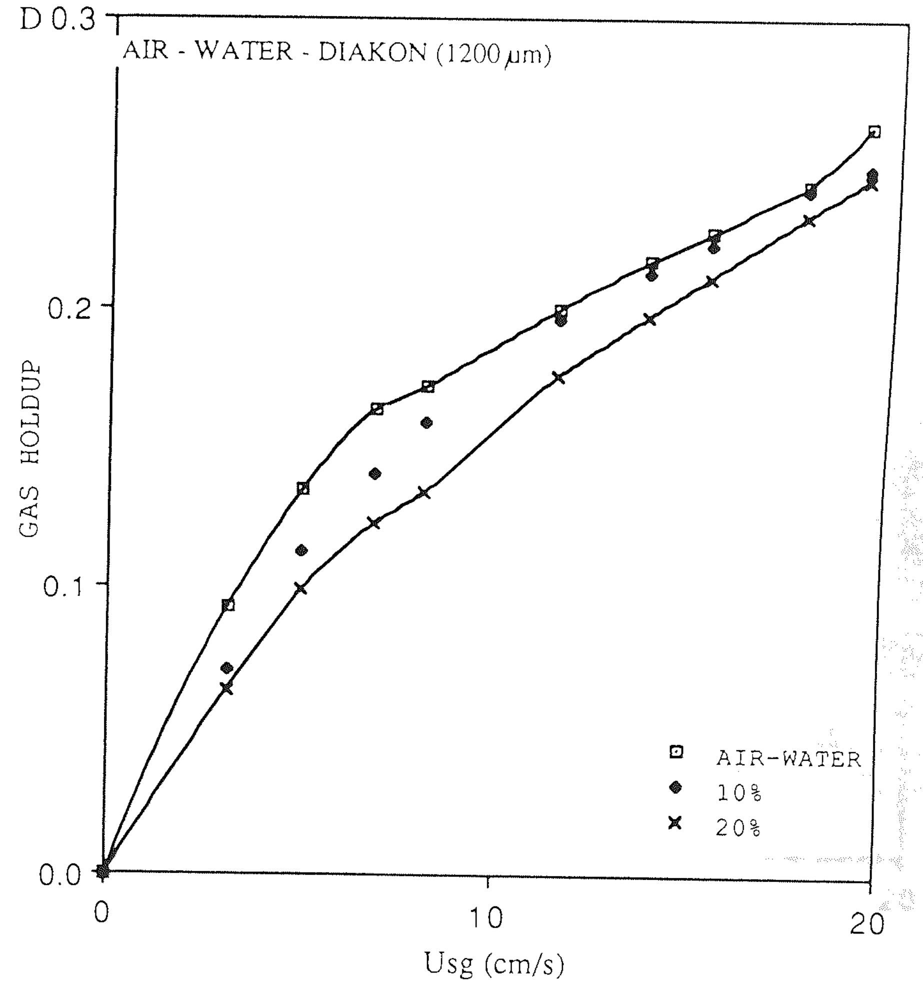


Figure 5.2 continued

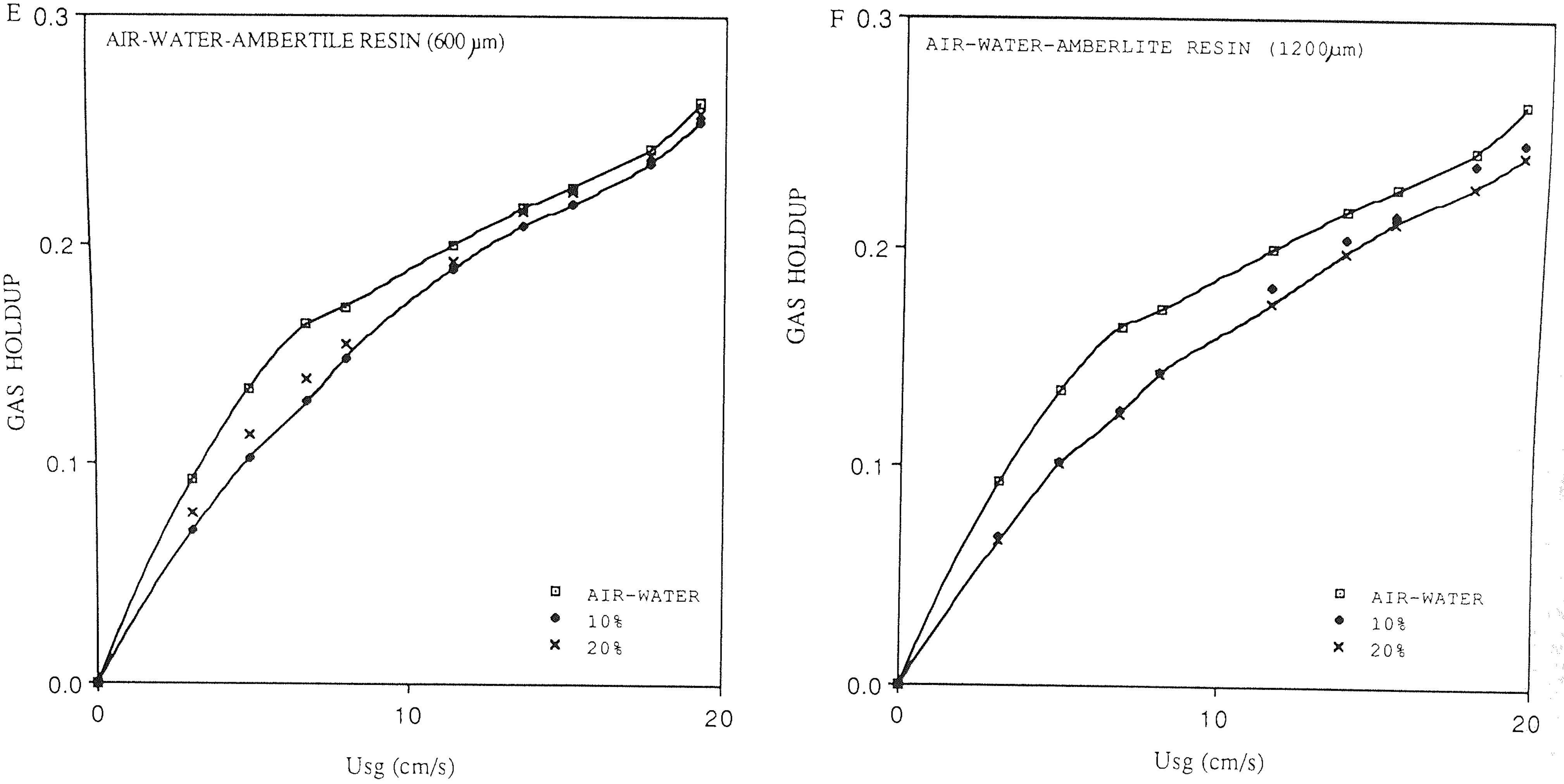


Figure 5.3: Effect of solid concentration on gas holdup in a 3D column

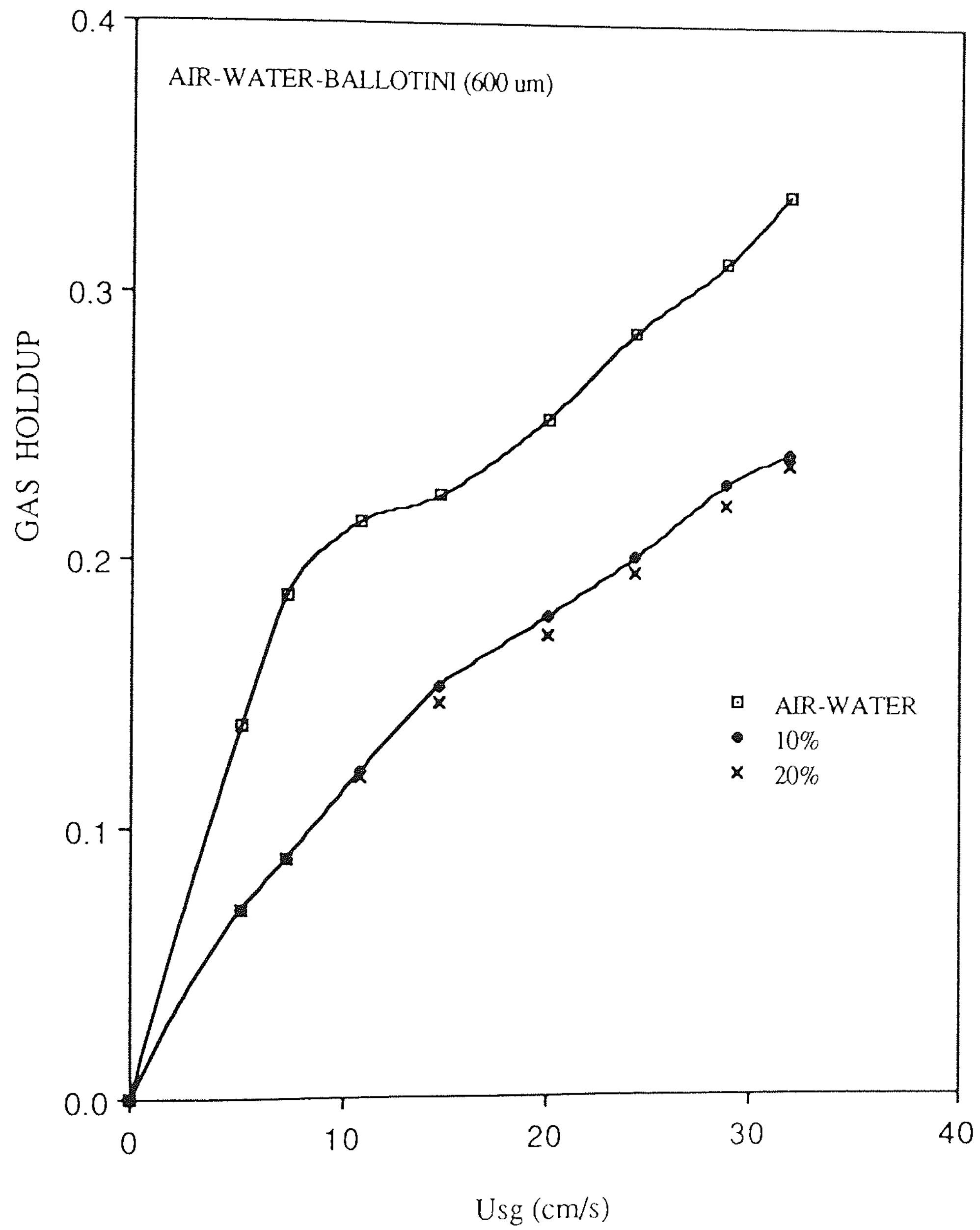
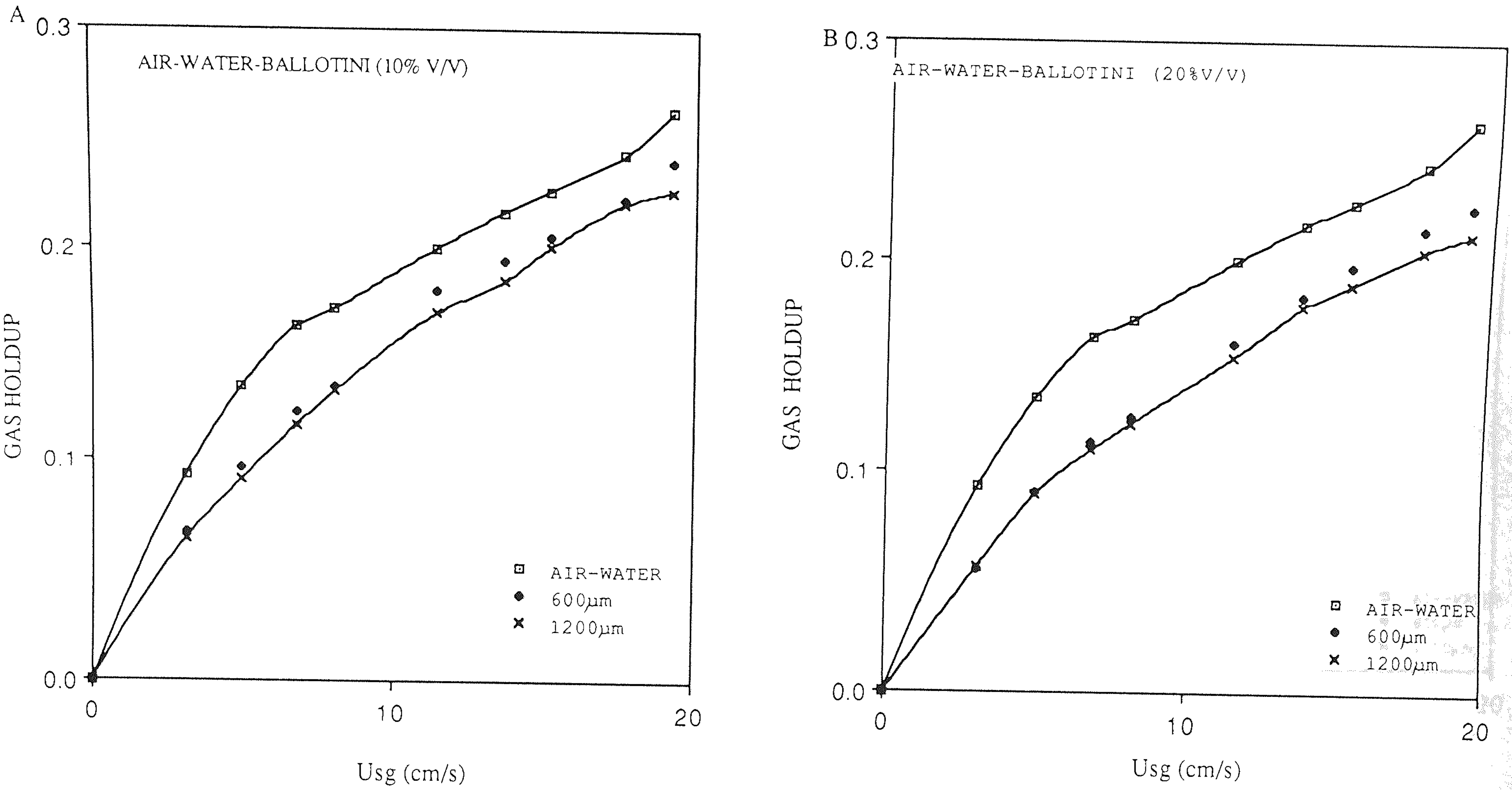
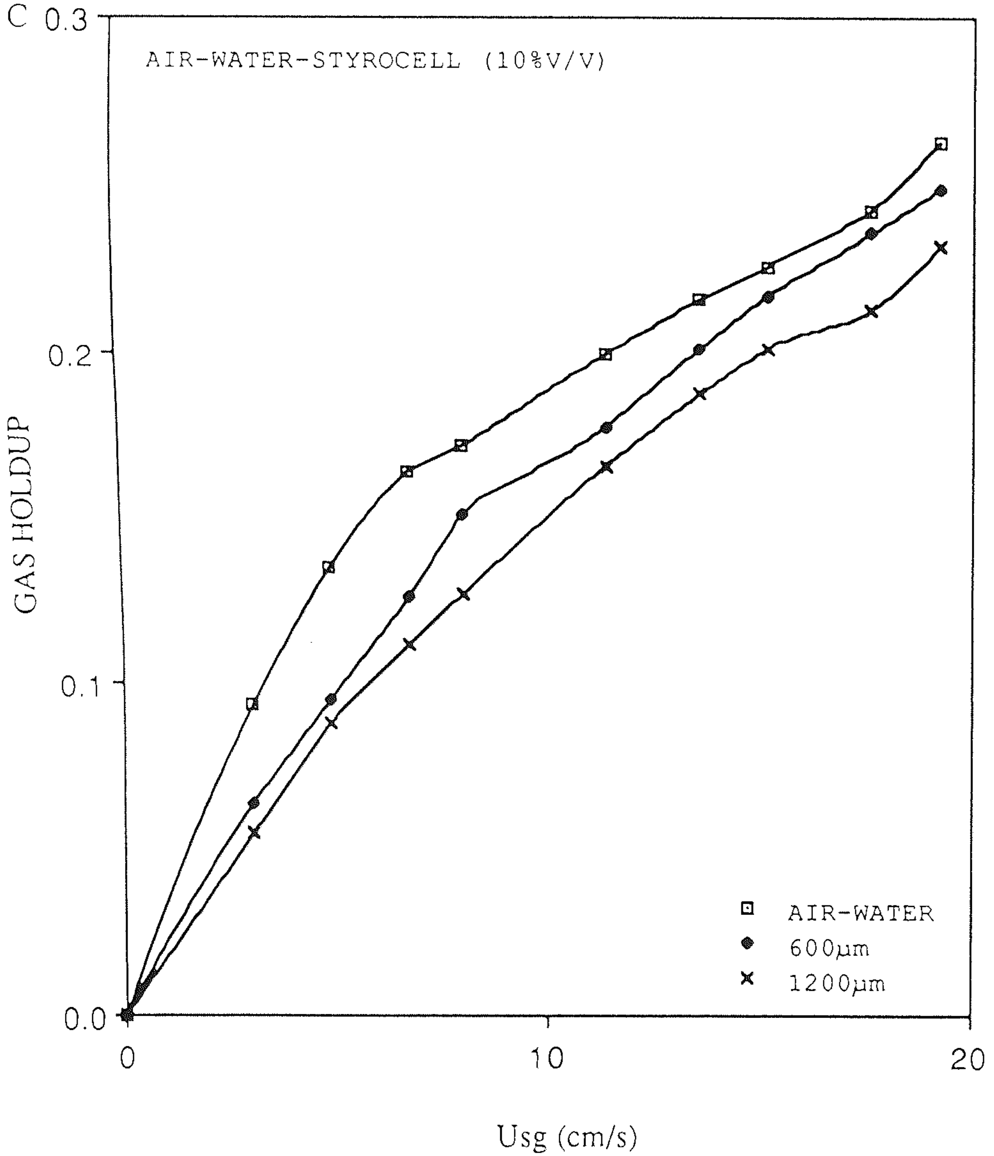


Figure 5.4 : Effect of solid size on gas holdup (2D bubble column)




C 0.3
AIR-WATER-STYROCELL (10%V/V)
GAS HOLDUP
0.2
0.1
0.0
0
10
20
Usg (cm/s)
AIR-WATER
600μm
1200μm

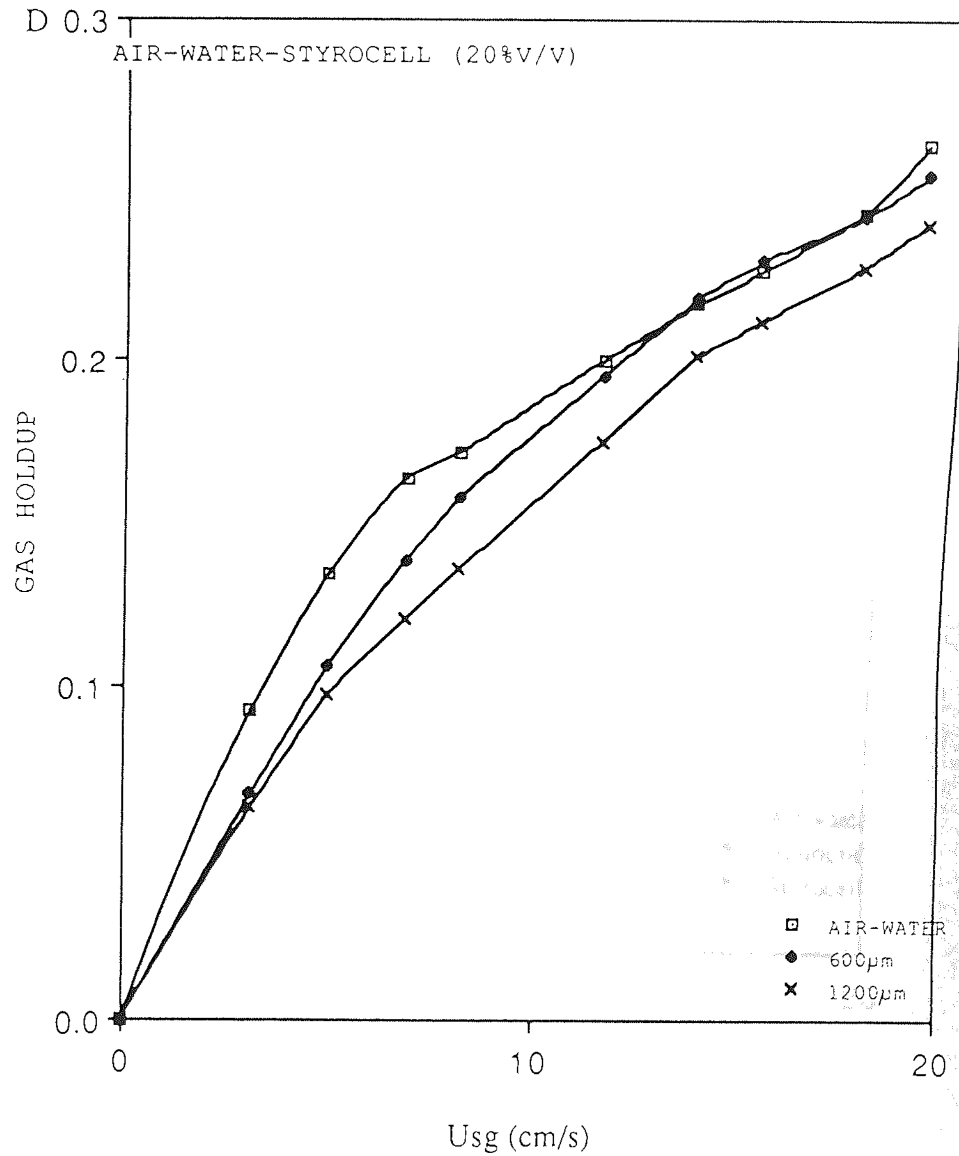
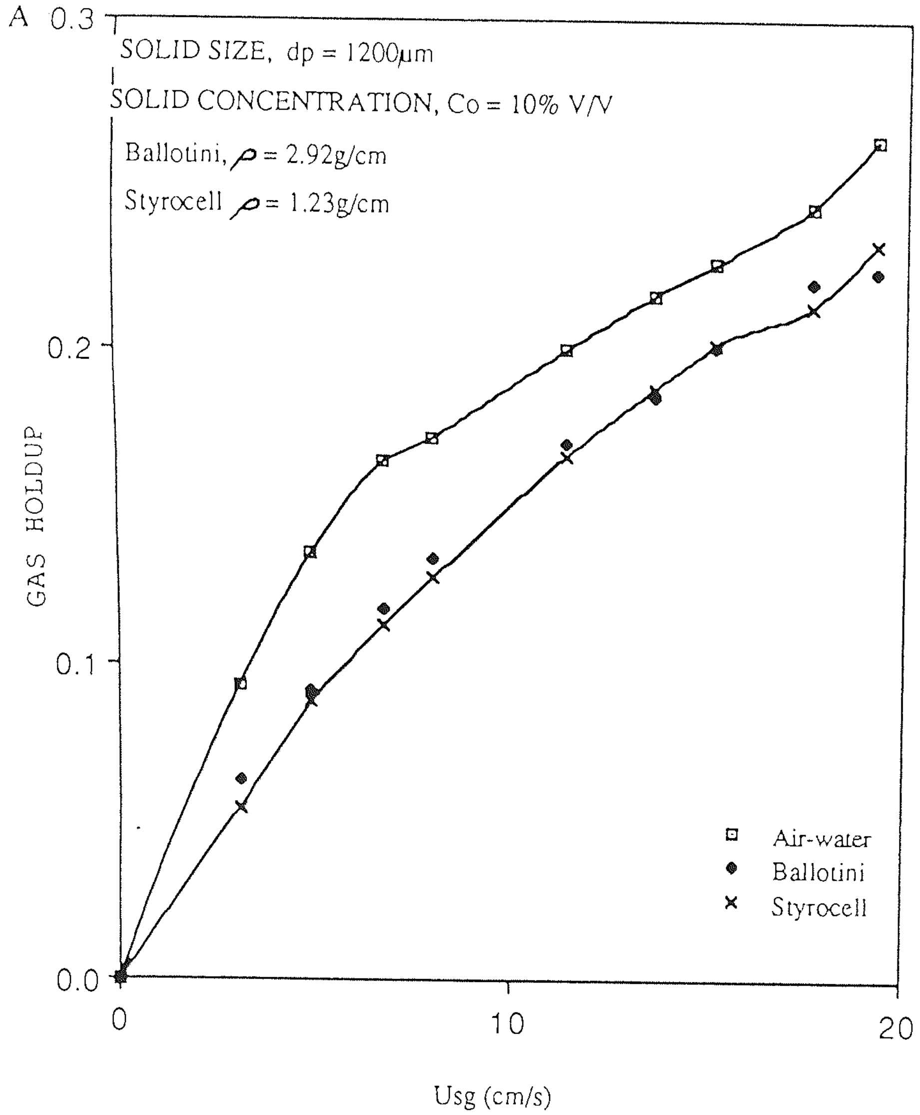
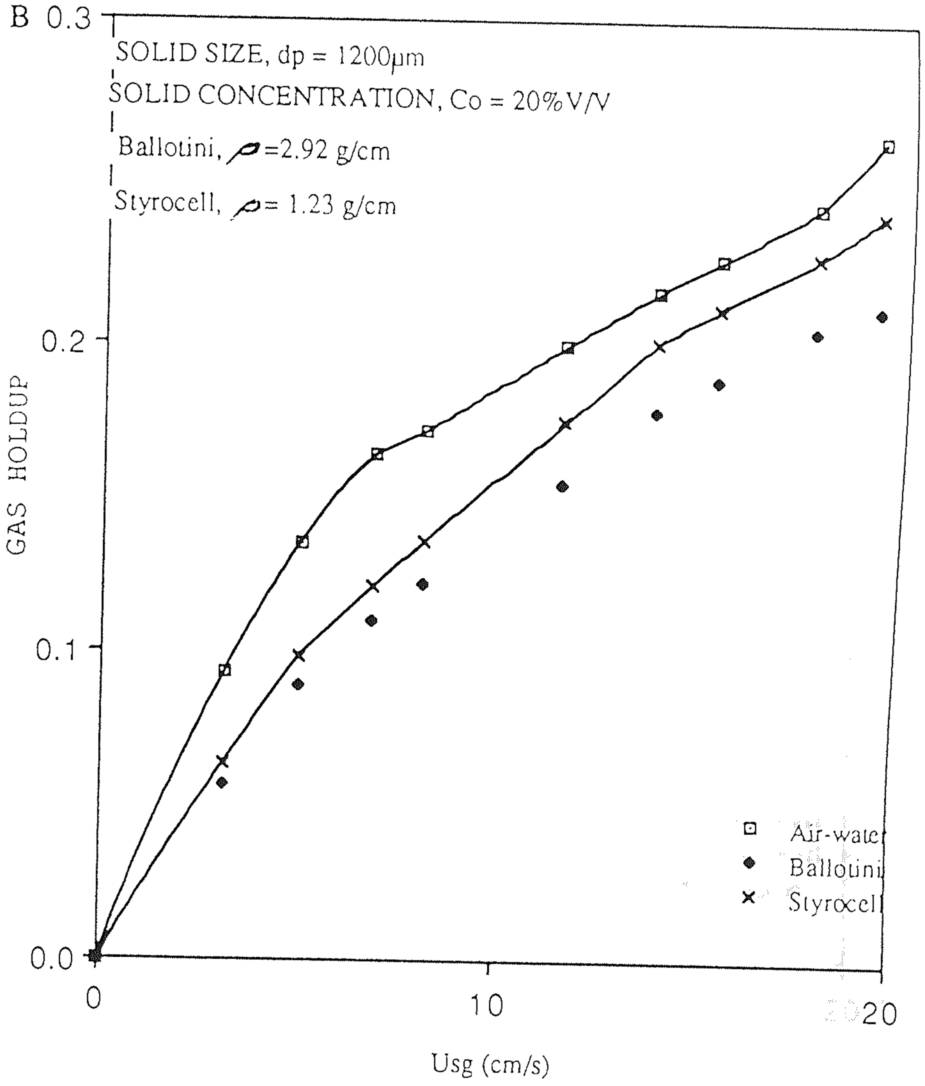
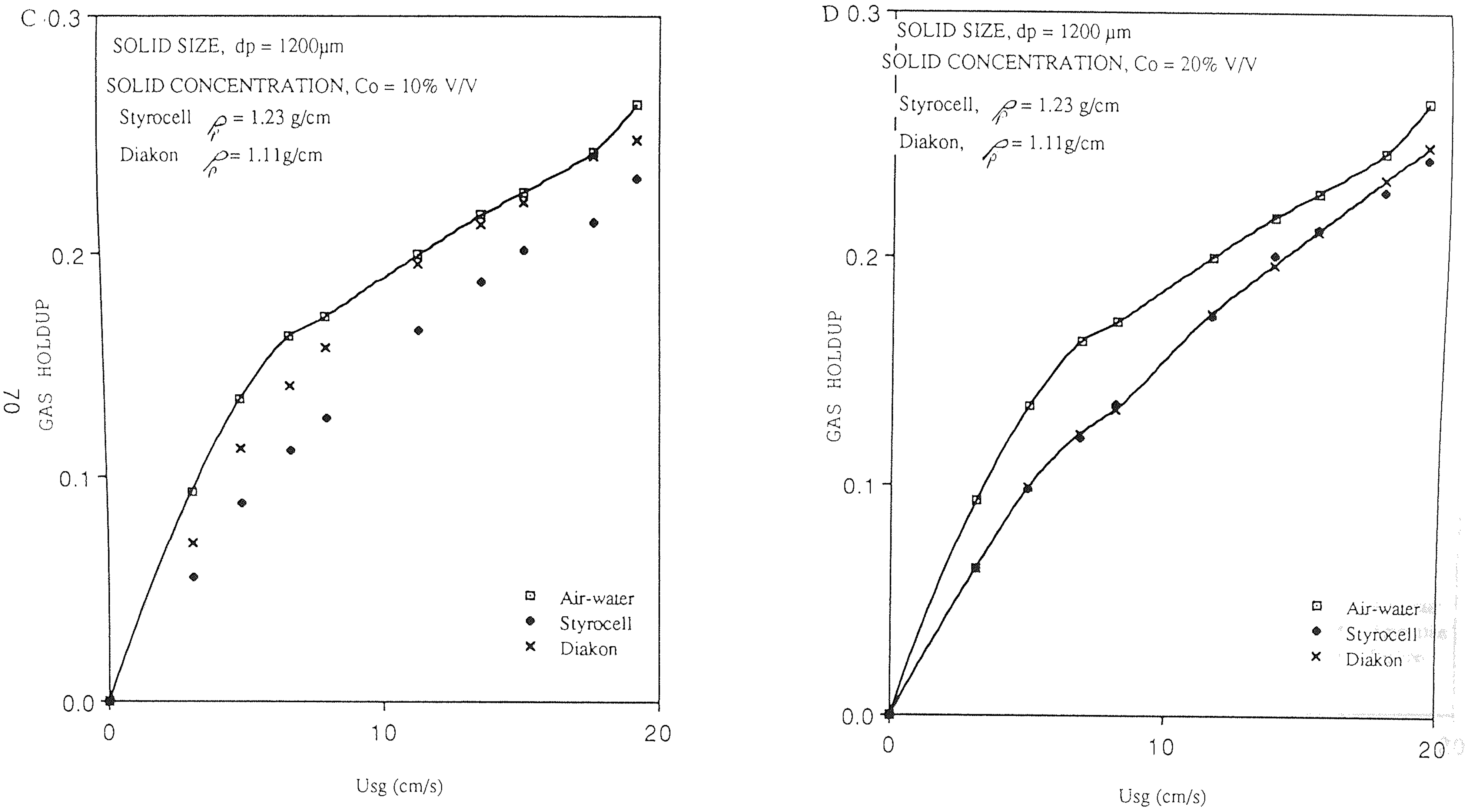
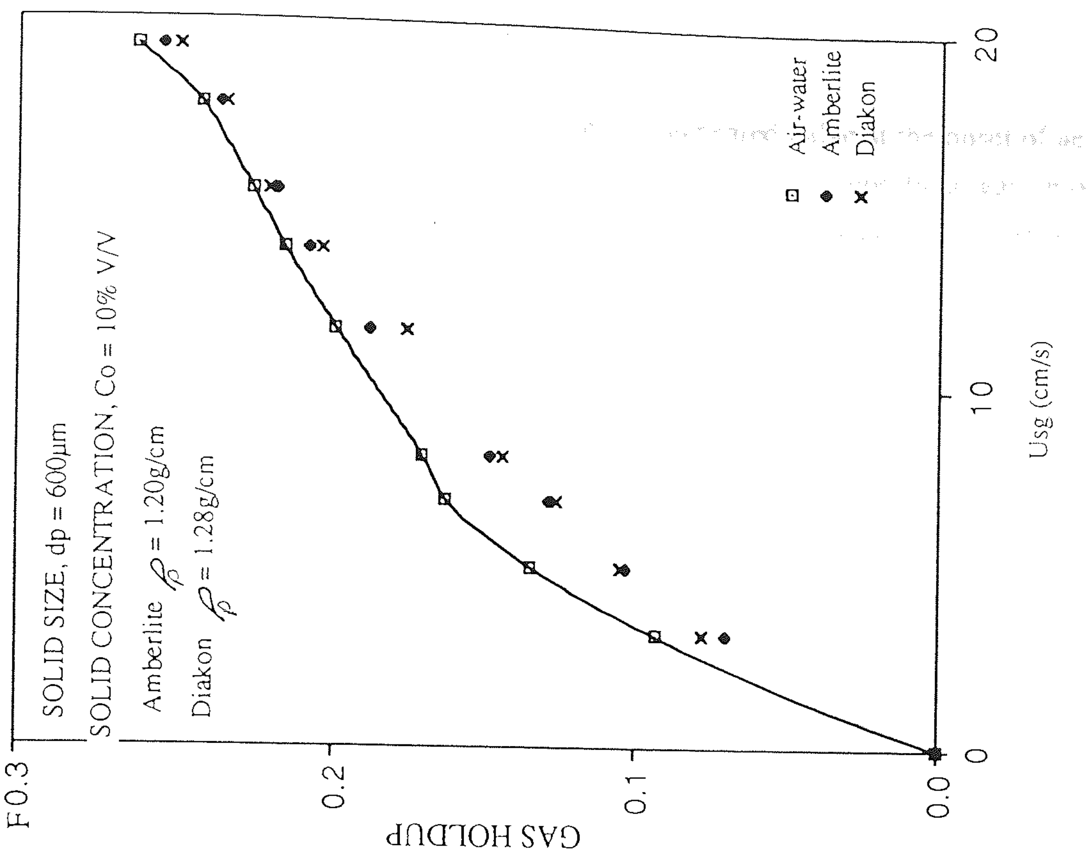

D 0.3
AIR-WATER-STYROCELL (20%V/V)
GAS HOLDUP
0.2
0.1
0.0
0
10
20
Usg (cm/s)
AIR-WATER
600μm
1200μm

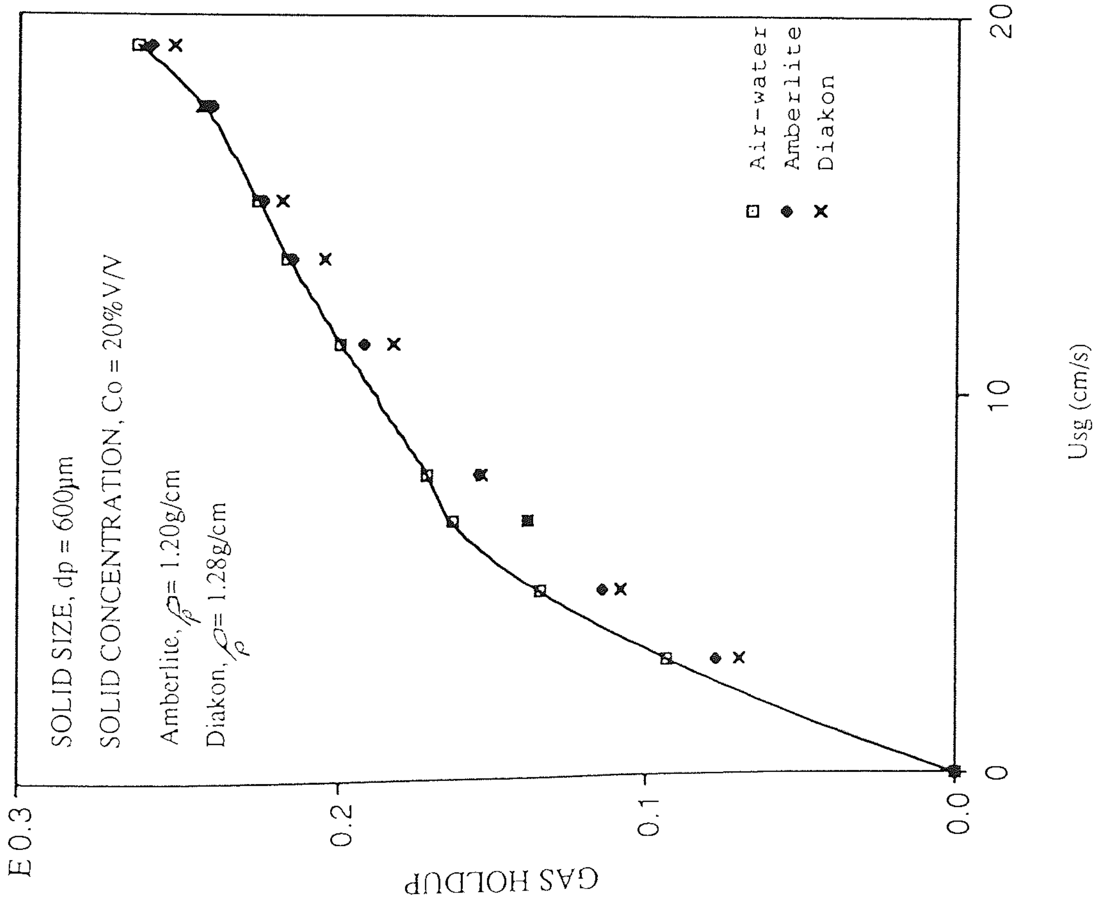
Figure 5.5 : Effect of solid density on gas holdup






C 0.3
SOLID SIZE, dp = 1200μm
SOLID CONCENTRATION, Co = 10% V/V
Styrocell ρ = 1.23 g/cm
Diakon ρ= 1.11g/cm
GAS HOLDUP
0.2
0.1
0.0
0
10
20
Usg (cm/s)
Air-water
Styrocell
Diakon
D 0.3
SOLID SIZE, dp = 1200 μm
SOLID CONCENTRATION, Co = 20% V/V
Styrocell, ρ= 1.23 g/cm
Diakon, ρ= 1.11g/cm
GAS HOLDUP
0.2
0.1
0.0
0
10
20
Usg (cm/s)
Air-water
Styrocell
Diakon


F
SOLID SIZE, dp = 600μm
SOLID CONCENTRATION, Co = 10% V/V
Amberlite ρ = 1.20g/cm
Diakon ρ = 1.28g/cm
Air-water
Amberlite
Diakon
GAS HOLDUP
0.0
0.1
0.2
0.3
0
10
20
Usg (cm/s)


E
SOLID SIZE, dp = 600μm
SOLID CONCENTRATION, Co = 20% V/V
Amberlite, ρ = 1.20g/cm
Diakon, ρ = 1.28g/cm
Air-water
Amberlite
Diakon
GAS HOLDUP
0.0
0.1
0.2
0.3
0
10
20
Usg (cm/s)

5.3.2 SOLID HOLDUP

The overall solid concentration distribution appeared halve at the onset of aeration; as expected, holdup gradually decreased with an increase in superficial gas velocity to an almost constant solid holdup value. Figure 5.6 shows typical plots of the solid holdup obtained for air-water-Ballotini (1200μm) at nominally 10% v/v initial solid concentration in 2D and 3D columns. The holdup of solid near the distributor (4cm above distributor plate) was found to be uncharacteristicall higher than in the rest of the columns.

Overall solid holdup reduced with increasing superficial liquid velocity at any axial position along the column length.

EFFECT OF SOLID CONCENTRATION

Figure 5.7 show typical results obtained for all the four particles in batch operation. Experimental data are listed in appendix B7.

Increasing the initial concentration of the total system only produced a shift in the solid holdup variation with superficial gas velocity.

EFFECT OF PARTICLE SIZE

Figure 5.8 shows the effect of particle size on the solid holdup. Very little effect was observed for systems except the air-water-Diakon one, in which increasing the particle size led to a noticeable increase in average solid holdup.

EFFECT OF PARTICLE DENSITY

Looking at figure 5.7 and 5.8 again, particles of similar densities have identical solid holdup characteristics. When comparing systems containing Ballotini with those containing Styrocell, higher solid holdup values were obtained with ballotini which was of higher particle density.

Figure 5.6: Effect of superficial gas and liquid velocities on solid holdup

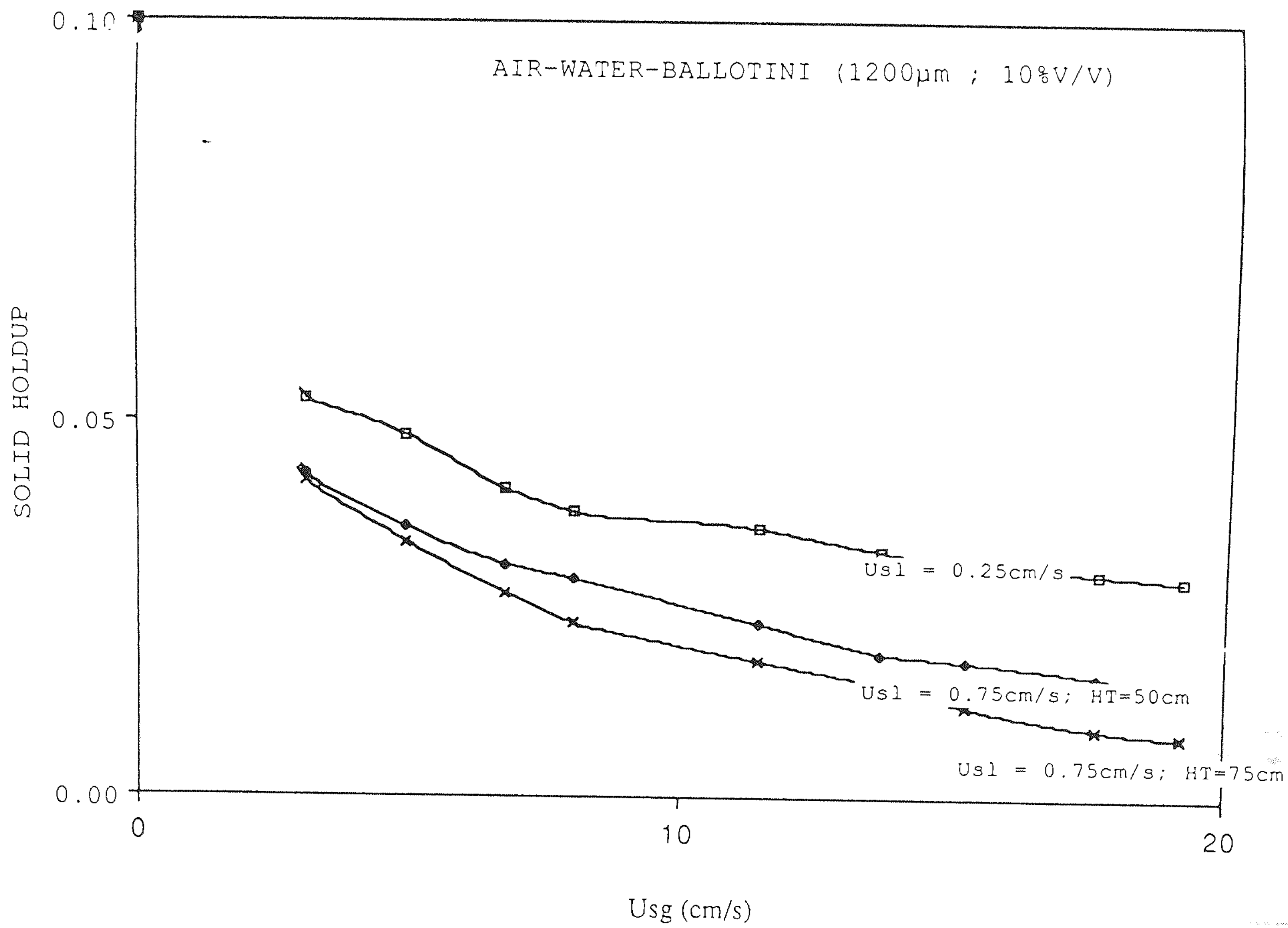


Figure 5.7 : Effect of solid concentration on solid holdup

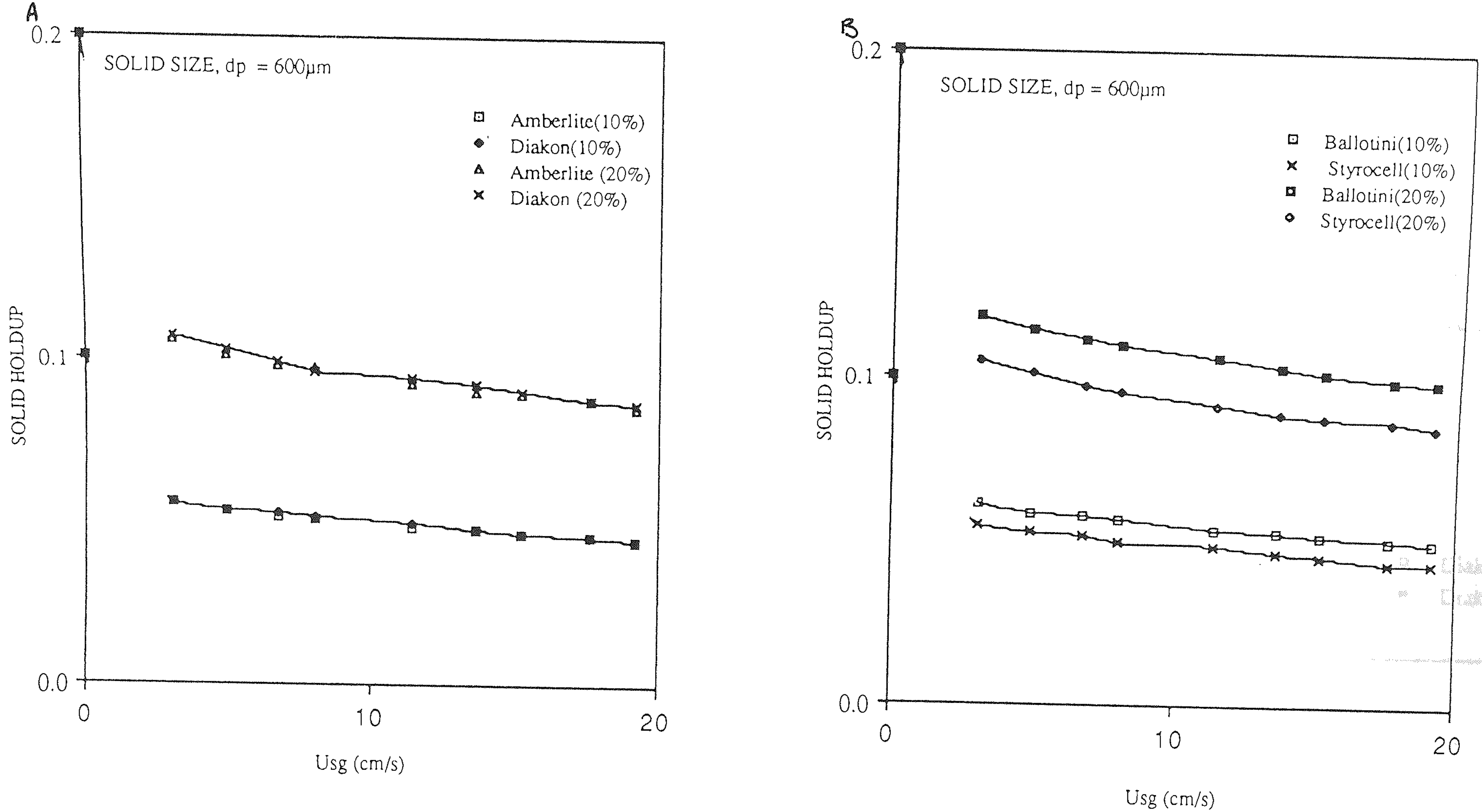
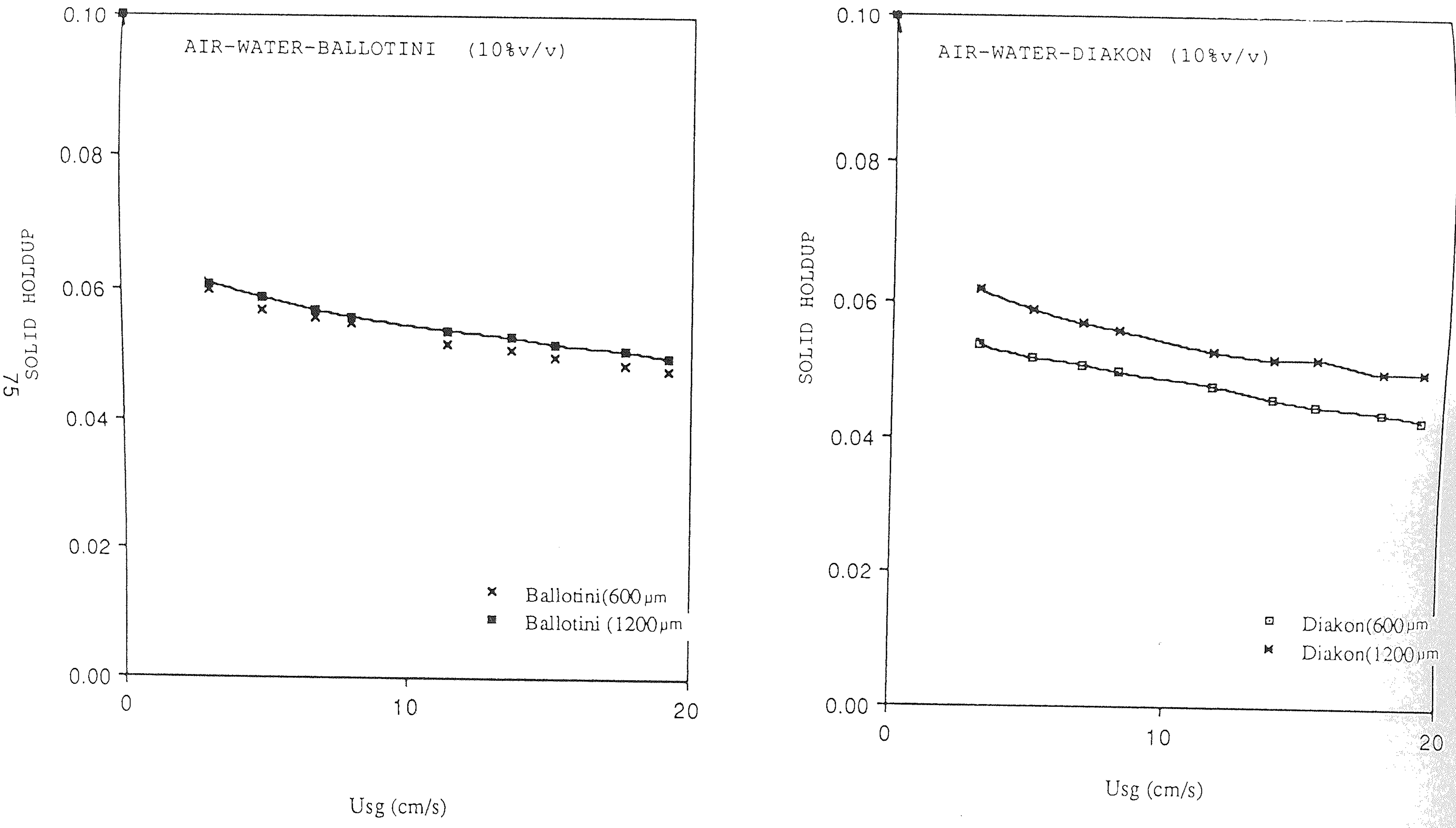


Figure 5.8 Effect of particle size on solid holdup



CHAPTER SIX

PHASE HOLDUP IN THREE-PHASE FLUIDISED SYSTEMS CONTAINING BINARY PARTICLE MIXTURES

6.1 INTRODUCTION

Overall gas and solid holdup characteristics were investigated in cocurrent gas-liquid-solid fluidised bubble columns containing binary particle mixtures. The binary mixtures were made up of four basic types of near spherical particles differing in size and/or density. The effect of the total mixture concentration, relative particle concentration, particle size ratio and density ratio on the phase holdups are presented.

Very limited information is available in the literature regarding phase holdup characteristics in three-phase heterogeneous fluidised beds with mixed size and/or density particles. Binary particle mixtures of this nature are often used in the study of mixing and segregation phenomena (see also chapter 9). For this reason, the review of the literature relevant to this section was conducted in connection with that supporting the mixing and segregation studies and as presented in chapter 3.

Axial variations of the holdups are discussed in chapter 9 in connection with the mixing and segregation studies. Comparisons with the behaviour of two phase systems and systems containing single component particles are reported in chapter 10.

6.2 EXPERIMENTAL PROCEDURE

Experiments were performed in the two- and three-dimensional bubble columns (see section 4.4.1 and 4.4.2). Binary particle mixture properties in terms of the three volume ratios 1:1, 1:2, and 2:1 were investigated in a systematic order at two concentration levels, 10% and 20% volume/volume. The physical properties of the solid particles have been presented in Tables 4.1 and 4.2 in chapter 4. The binary mixtures used in the studies are summarised in Table 6.1 below. Operating procedures as discussed in chapter 4 were followed. The particle binary mixtures were pre-mixed before introduction into the bubble columns.

The evaluation of the holdup of the individual phases was obtained from the relationships discussed in chapter 2.

For a system containing a binary particle mixtures, the solid holdup was evaluated from the relationships given below:

$$\varepsilon_S = (w_1/\rho_{s1} + w_2/\rho_{s2}) / A_{cs}H_f \qquad 6.1$$

$$-dp/dz = [\varepsilon_G \rho_G + \varepsilon_L \rho_L + (\varepsilon_{s1} \rho_{s1} + \varepsilon_{s2} \rho_{s2})] g \qquad 6.2$$

Table 6.1: Description of binary mixture of particles used

Mixture Notation	Description	Volume Ratio (Large/small)	Size Ratio	Weight Ratio	Density Ratio (Heavy/ Light)
M1a	Ballotini/Styrocell	1 : 1 1 : 2 2 : 1	1 : 1	2.9 1.4 5.4	2.6 ; 2.4
M1b		1 : 1 1 : 2 2 : 1	1: 2	2.9 1.4 5.4	2.3
M1c		1 : 1 1 : 2 2 : 1	2 : 1	3.1 1.5 5.8	2.3
M2a	Ballotini/Diakon	1 : 1 1 : 2 2 : 1	1 : 1	2.4 1.3	2.2
M2b		1 : 1 1 : 2 2 : 1	1 : 2	2.4 1.3	2.6
M2c		1 : 1 1 : 2 2 : 1	2 : 1	2.6 1.3 5.2	2.6
M3a	Ballotini/Amberlite	1 : 1 1 : 2 2 : 1	1 : 1	2.85	2.4
M3b		1 : 1 1 : 2 2 : 1	1 : 2	1.4 5.5	2.5
M3c		1 : 1 1 : 2 2 : 1	2 : 1	1.1 1.4 5.4	2.5
M4a	Styrocell/Diakon	1 : 1 1 : 2 2 : 1	1 : 1		1.15
M4b		1 : 1 1 : 2 2 : 1	1 : 2	1.00 1.96 1.60	

Table 6.1 continued

M4c		1 : 1	2 : 1		1.00
		1 : 2		2.10	
		2 : 1		1.76	
M5a	Styrocell/Amberlite	1 : 1	1 : 1		1.08
		1 : 2			
		2 : 1			
M5b		1 : 1	1 : 2		1.02
		1 : 2		2.2	
		2 : 1		2.03	
M5c		1 : 1	2 : 1		1.02
		1 : 2		2.75	
		2 : 1		2.01	
M6a	Diakon/Amberlite	1 : 1	1 : 1	1.03	1.07
		1 : 2			
		2 : 1			
M6b		1 : 1	1 : 2		1.13
		1 : 2		1.8	
		2 : 1			
M6c		1 : 1	2 : 1		1.13
		1 : 2		1.97	
		2 : 1		2.07	

and

$\varepsilon_g + \varepsilon_L + \varepsilon_s = 1$ 6.3

The overall gas holdup was evaluated from the expanded bed height as in the case of air-water and systems containing single components using equation 5.1

(i.e., ε_g = Hf - Ho/Hf)

A comparison and discussion of the results is given in chapter 10.

6.3 EXPERIMENTAL RESULTS

Tables of the experimental results from which the graphs were prepared are provided in appendix C.

6.3.1 GAS HOLDUP

The gas holdup trends obtained were similar to those observed for air-water and three phase systems containing single particles. However, the presence of a binary particle mixture appeared to cause a greater reduction in gas holdup as the superficial gas velocity increased. Unlike systems containing single particles, there was no condition under which the gas hold-up approached that for air-water.

EFFECT OF BINARY MIXTURE CONCENTRATION AND CONCENTRATION RATIO

(1) TOTAL CONCENTRATION

Two total binary mixture concentration levels were used -10% and 20% packed volume/volume, of the total system. Figure 6.1 illustrates the results obtained for two systems containing binary mixture particles - M1a (Ballotini/Styrocell, density ratio 2.4) and M6a (Diakon/Amberlite, density ratio 1.02) of equal size and binary concentration ratio. This was typical of results obtained for other cases.

On increasing the total concentration of the binary particle mixture from 10% to 20%, the reducing effect caused by the presence of the solids was diminished, particularly with binary particle mixture M1a (Ballotini/Styrocell) at high superficial gas velocities.

Figure 6.1: Effect of concentration on gas holdup in a system containing binary particle mixture

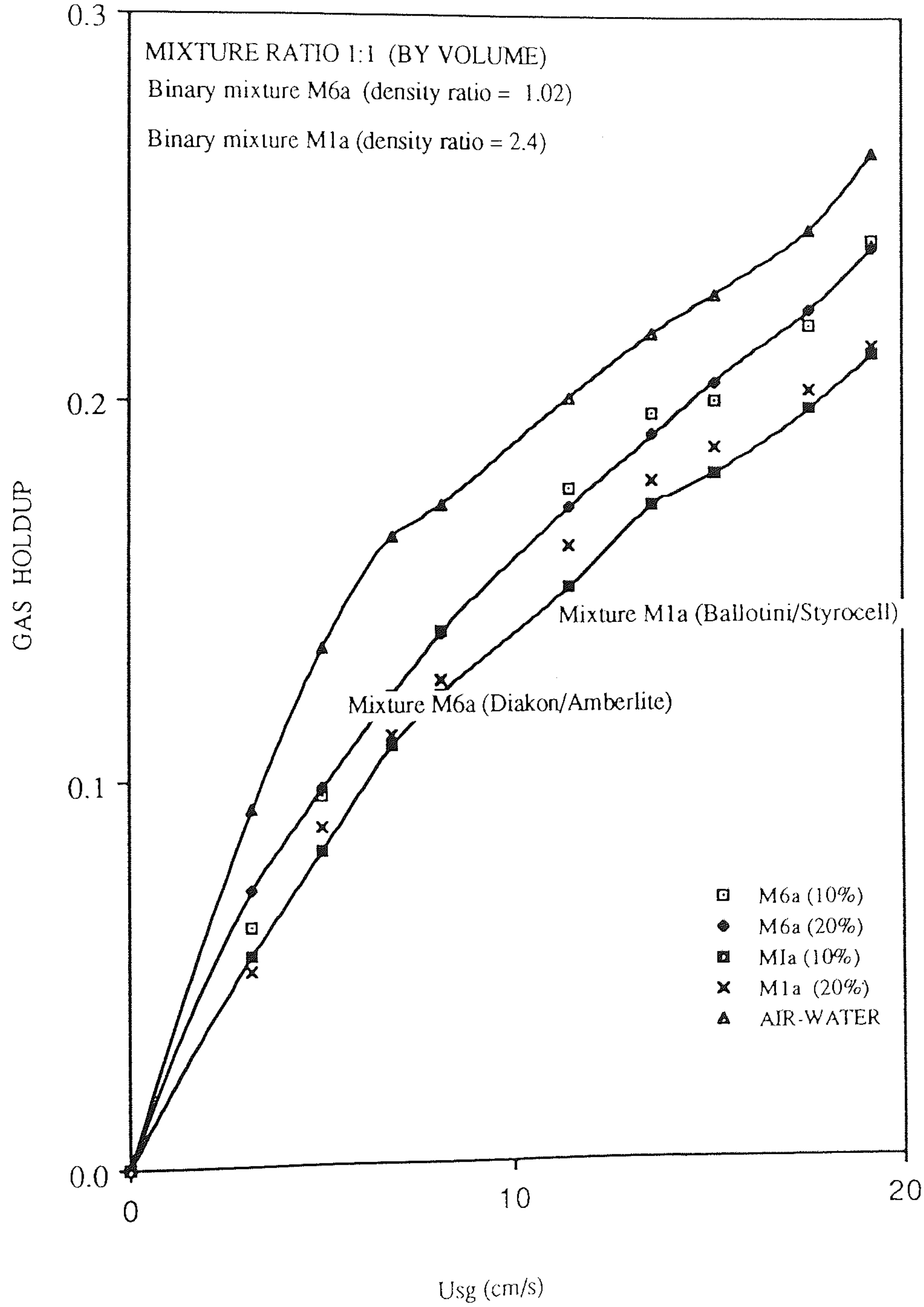
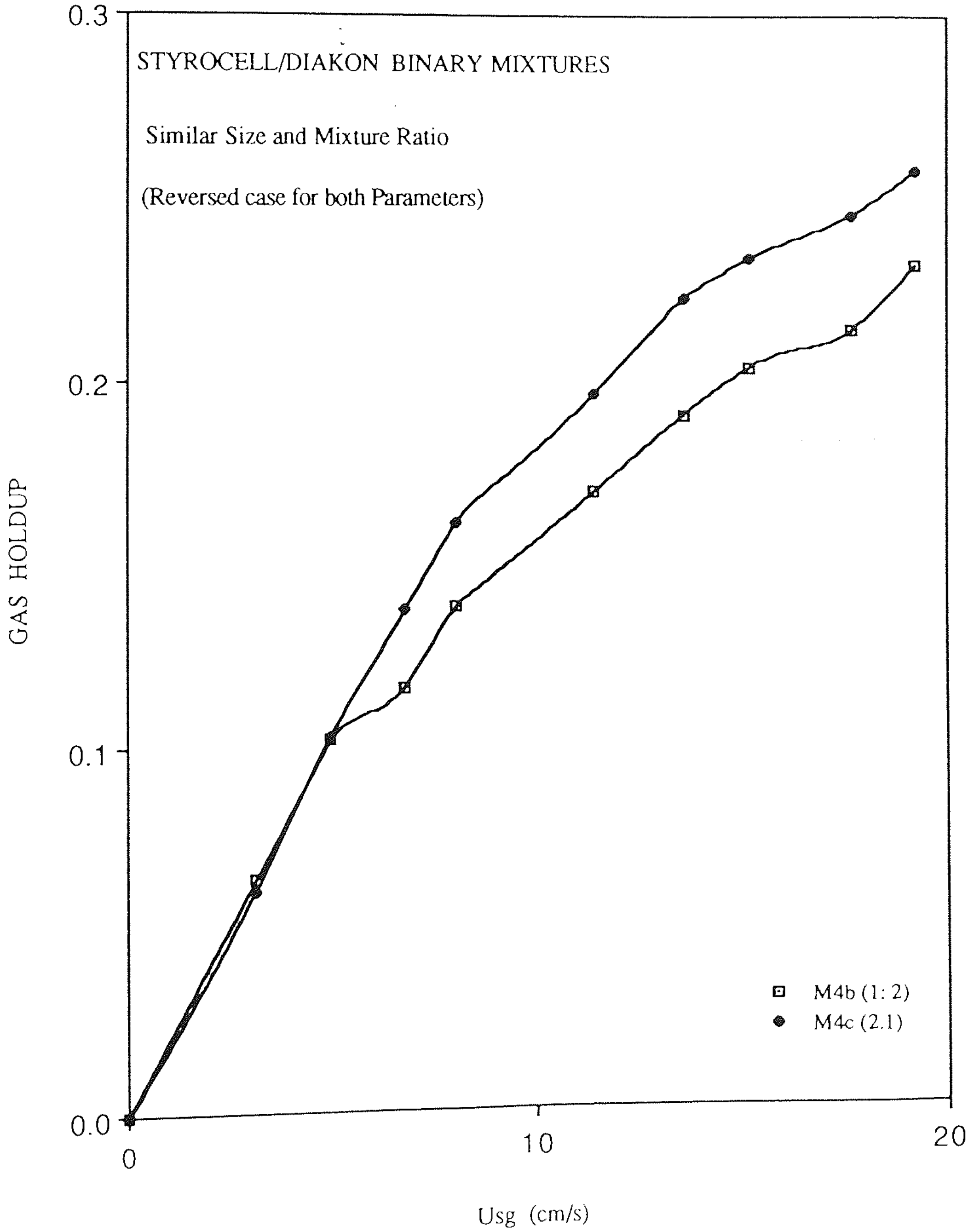


Figure 6.2: Effect of concentration ratio on gas holdup in a system containing binary particle mixture



Comparing with air-water, the effect of concentration did not produce any diminishing effect and trend towards that of air-water in either case.In contrast, systems containing single particles (chapter 5) particularly for Diakon, Amberlite or Styrocell which have densities close to that of water, showing identical trend, due to high degree of phase interraction. Please notte that oncentration ratio and volume ratio are equivalent.

(2) CONCENTRATION RATIO

Figure 6.2 shows the effect of concentration ratio for the binary mixtures M4b and M4c (Styrocell/Diakon). Individual particle components propertiess appeared to be interrraction with each other and the system: this might be thought of as some sort of "trade-off" of properties. The effect is only apparent when the size and mixture ratio are similar. When Styrocell particles were the major component, there was a clear reduction in gas hold-up at high superficial gas velocities.

EFFECT OF SIZE AND SIZE RATIO

Figure 6.3 illustrates the effect of size and size ratio on the gas holdup for three binary mixtures when present in equal proportion. Similar values were obtained for the size ratios 1: 1 and 2 : 1: when the ratio was 1: 2 marginally higher gas hold-ups were recorded. Looking back at figure 6.2, this was not surprising, as it highlights the competing nature of the physical parameters of the solids when present in a mixture. Tables C3 in the appendix C provide experimental data for other binary mixtures.

EFFECT OF DENSITY AND DENSITY RATIO

Tables C4 A and B in the appendix C provide lists of experimental data for the binary systems. Gas holdup values are very similar for particles having similar densities in the binary mixture.

Binary mixtures with different densities and density ratio brought about a noticeable reduction in gas holdup when present in equal proportions (see fig 6.4b). By contrast, Fig 6.4a shows plots illustrating that gas hold-up is not always strongly reduced in the presence of particles with high density ratio.

Figure 6.3: Effect of size and size ratio on gas holdup in a system containing binary particle mixtures

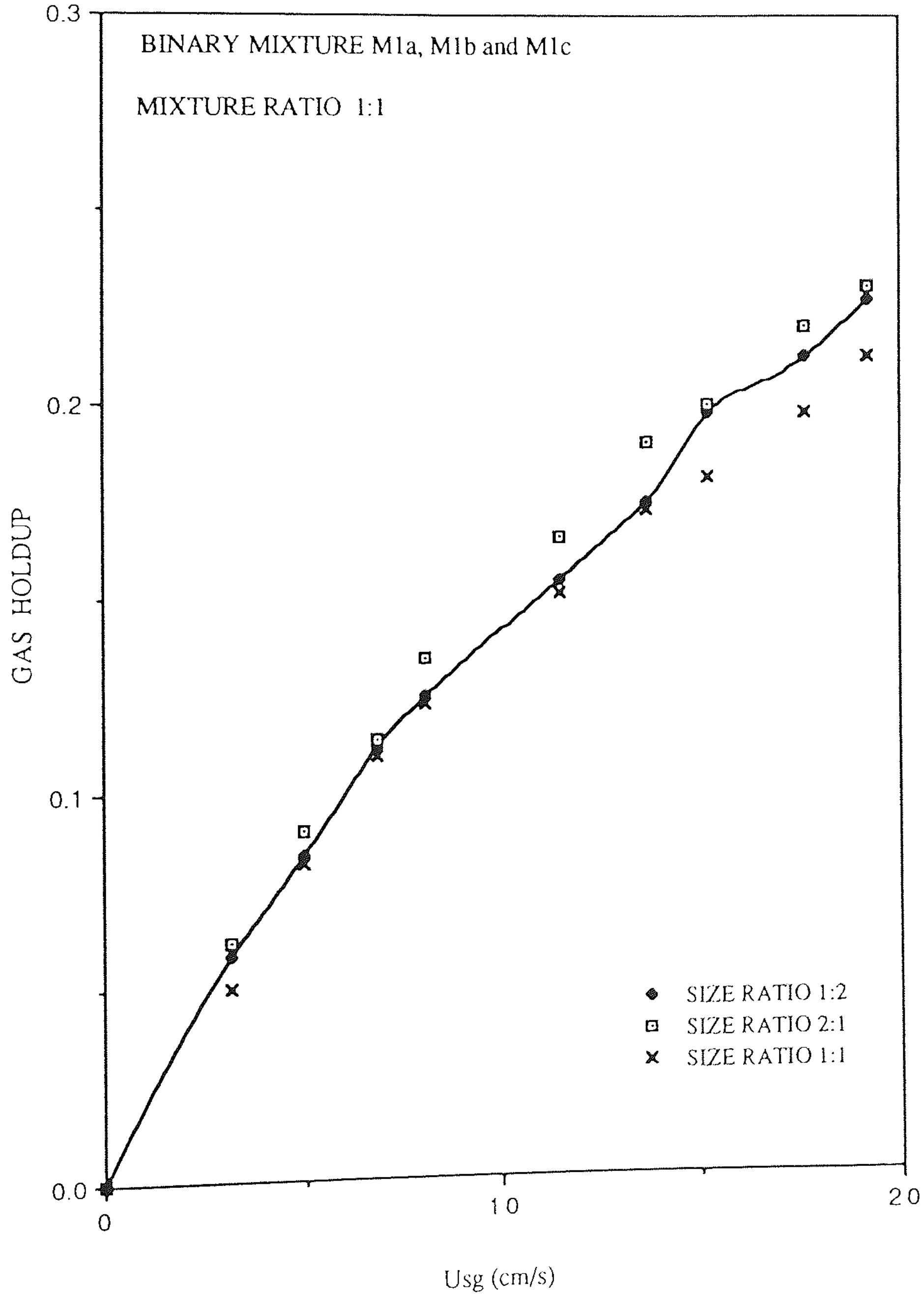
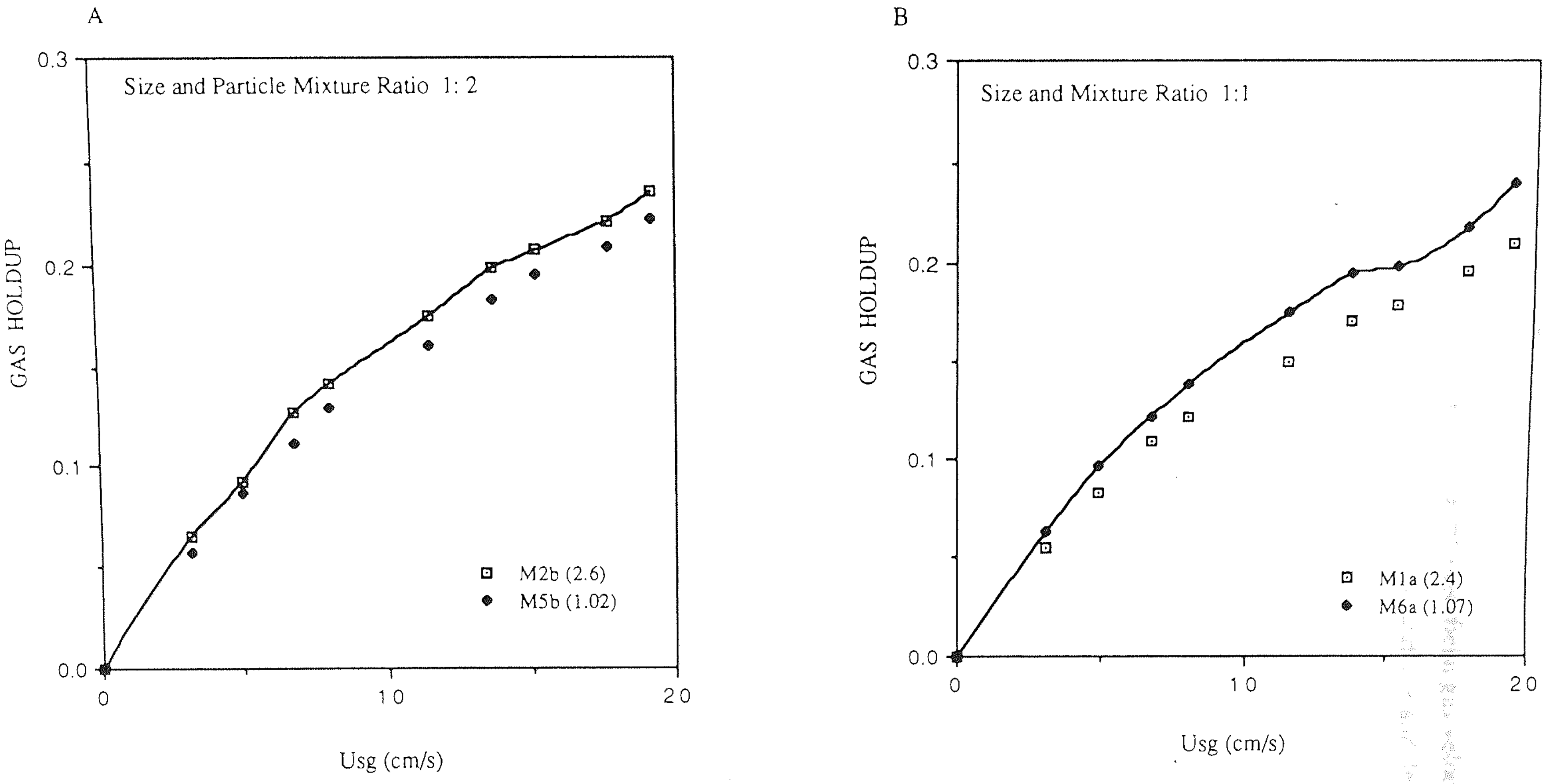


Figure 6.4: Effect of density and density ratio on gas holdup in a system containing binary particle mixtures



6.3.2 SOLIDS HOLDUP

The overall solids holdup reduced with increasing superficial gas velocity at any axial position irrespective of the binary mixture concentration and size and density ratios (see figure 6.5).

EFFECT OF
TOTAL CONCENTRATION AND CONCENTRATION RATIO

Figure 6.5 shows typical results obtained for all binary mixtures. Increasing the total concentration of the mixture only produces a shift in the holdup variation similar to those obtained from the systems containing single particles. However, the overall holdup contribution from each solid in the binary system was dependent on the concentration ratio, i.e individual concentration of each component of the binary mixture (see figure 6.6a and b).

EFFECT OF SIZE AND SIZE RATIO

Figure 6.7a and b show the effect of size and size ratio on solid holdup. Also, figure 6.6 illustrates the effect. Results presented in figure 6.6a and b were typical for the remaining binary mixtures.

EFFECT OF DENSITY AND DENSITY RATIO

From figures 6.5 and 6.6, the solid holdup is found to be sensitive to density and density ratio. Higher holdup values were obtained with binary systems containing Ballotini. Experimental values are listed in tables C5 and C6 (appendix C).

Figure 6.5: Effect of total concentration on solid holdup in a system containing binary particle mixture

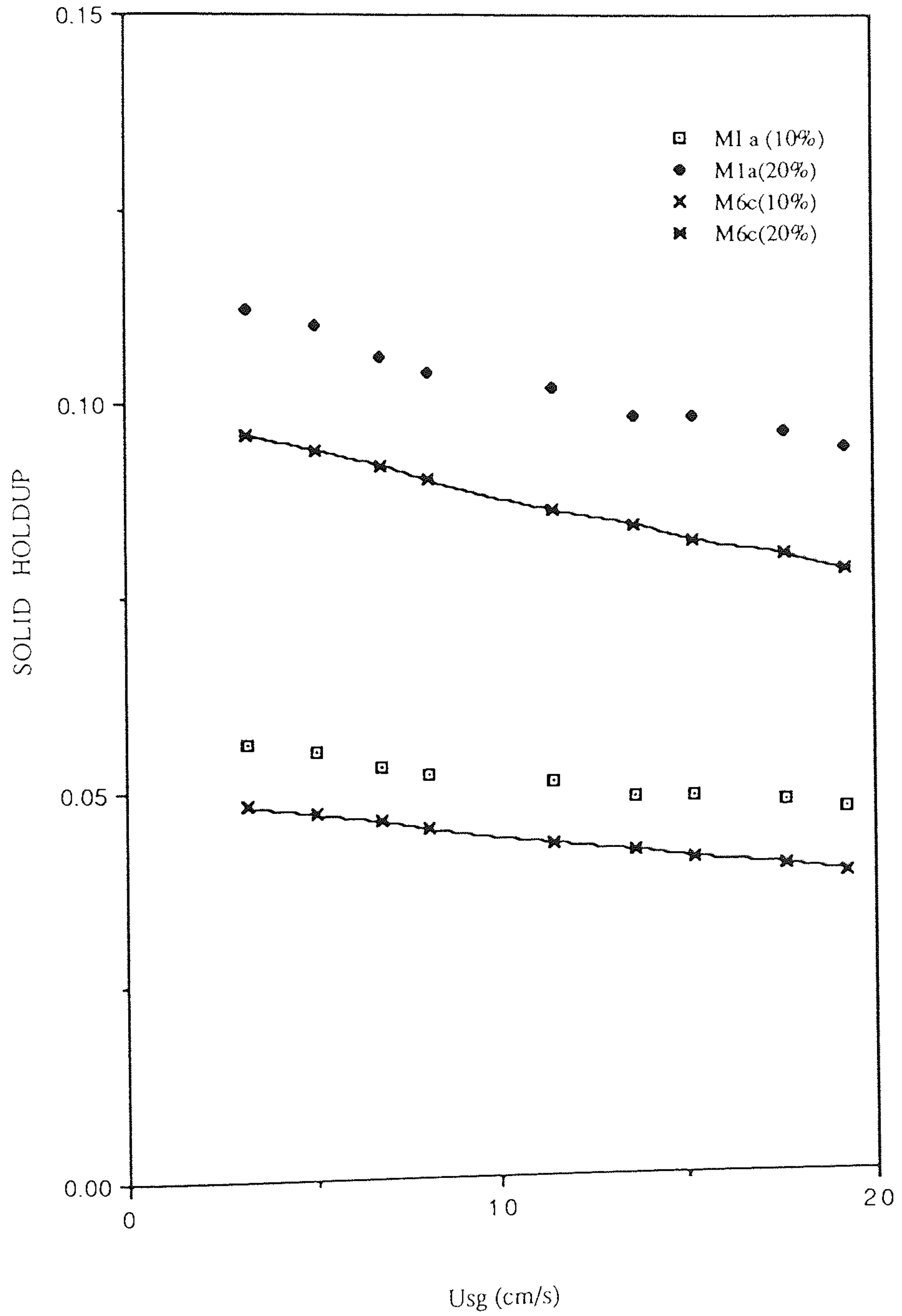
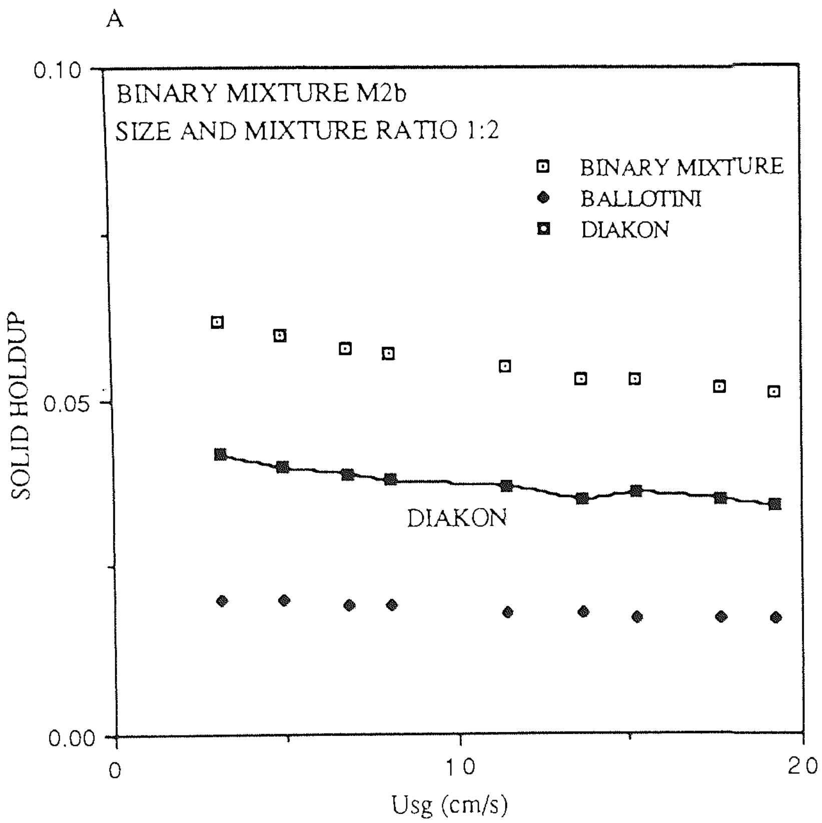


Figure 6.6: Effect of concentration ratio on solid holdup in systems containing binary particle mixture



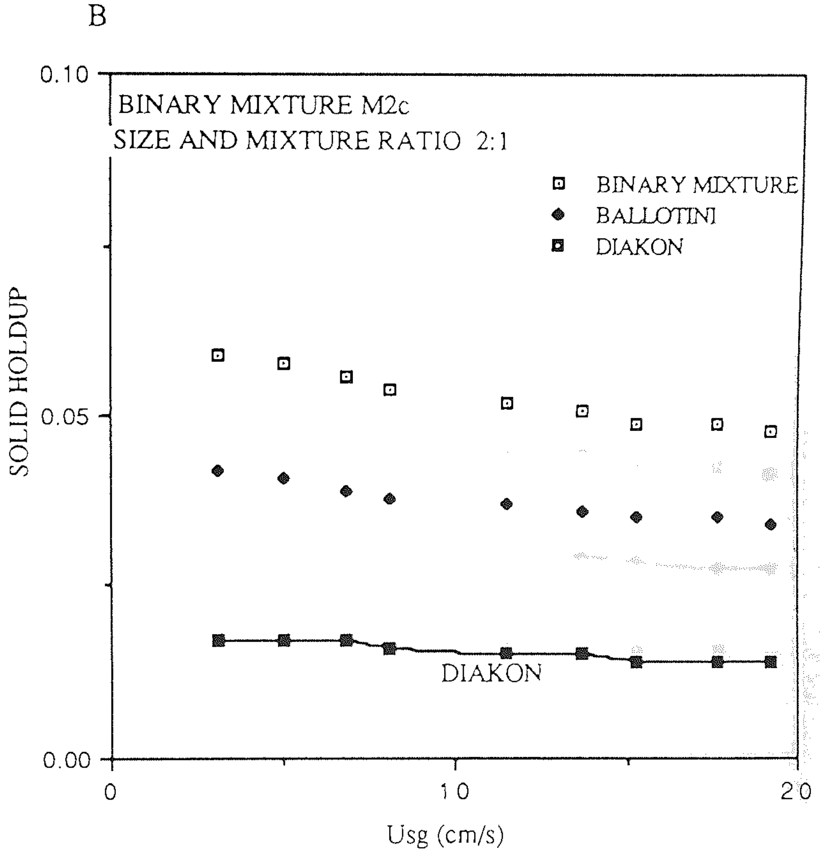
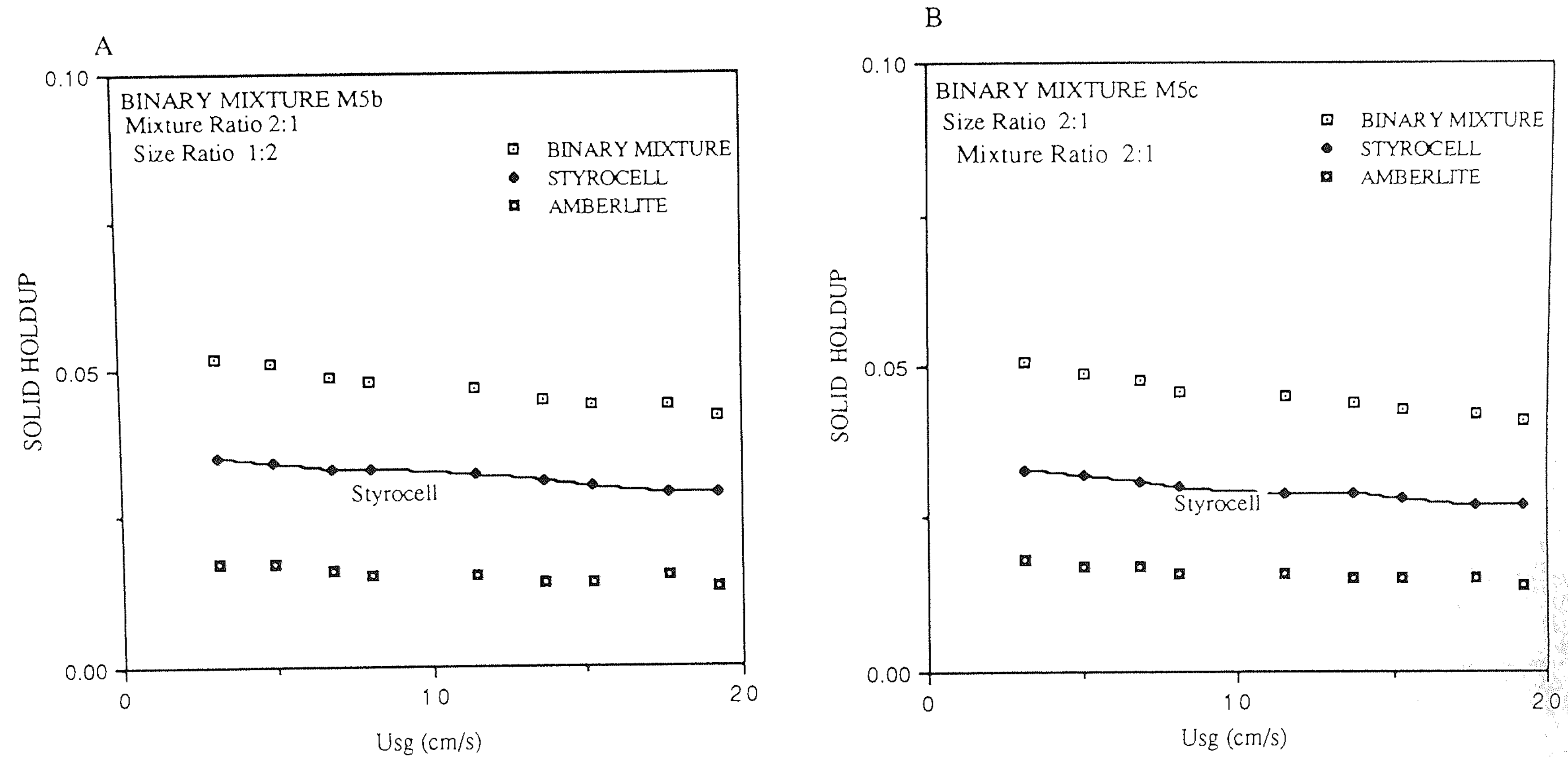


Figure 6.7: Effect of size and size ratio on solid holdup in systems containing binary particle mixture



CHAPTER SEVEN

PHASE HOLDUP IN THREE-PHASE FLUIDISED SYSTEMS CONTAINING TERNARY PARTICLE MIXTURES

7.1 INTRODUCTION

Overall gas and solid holdup characteristics were investigated in cocurrent complex gas-liquid-solid fluidised bubble columns containing ternary particle mixtures. The ternary mixtures were selected from the four basic types of near spherical particles, as used in chapter 6, differing in size and/or density. Comparison of results with those obtained in chapters 5 and 6 were made considering the total mixture concentration, relative particle concentration, particle size ratio and density ratio on the phase holdups. Because of small variations produced in the effects investigated, and, therefore to prevent duplication of similar graphs, the tables of results are only used and provided in the appendices, (Appendix D).

To the author's knowledge, no information is available in the literature regarding phase holdup characteristics in three-phase heterogeneous fluidised beds with mixed size and/or density particles of this nature. Due to difficulties in analysis of the complex system, experiments were limited to three mixtures at two concentration levels.

7.2 EXPERIMENTAL PROCEDURE

Experiments were performed in the two- and three-dimensional bubble columns (see section 4.4.1 and 4.4.2). Ternary particle mixture properties in terms of the three volume ratios 1:1:1, 1:1: 2, and 2:2:1 were investigated in a systematic order at two concentration levels, 10% and 20% volume/volume. The physical properties of the solid particles have been presented in Tables 4.1 and 4.2 in chapter 4. The ternary mixtures used in the studies are sumarised in Table 7.1 below. Operating procedures as discussed in chapter 4 were followed. The ternary particle mixtures were pre-mixed before introduction into the bubble columns.

As in chapter 6, the evaluation of the holdup of the individual phases was obtained from the relationships discussed in chapters 4 and 5.

For a system containing a ternary particle mixture, the solid holdup was evaluated from the relationships given below:

$$\varepsilon_S = (w_1/\rho_{s1} + w_2/\rho_{s2} + w_3/\rho_{s3}) / A_{cs}H_f \qquad 7.1$$

$$-dp/dz = [\varepsilon_G \rho_G + \varepsilon_L \rho_L + (\varepsilon_{s1} \rho_{s1} + \varepsilon_{s2} \rho_{s2} + \varepsilon_{s3} \rho_{s3})] g \qquad 7.2$$

Table 7.1 : Summary of the ternary particle mixtures used

Mixture Notation	Description	Volume Ratio	Size Ratio
MT1	Ballotini /Styrocell /Amberlite	1 : 1: 1	1 : 1: 1
MT2	Styrocell/Diakon/Amberlite	1 : 1: 2	2 : 2 :1
MT3	Ballotini/Styrocell/ Diakon	2 : 2: 1	1 : 1 : 2

and

$\varepsilon_g + \varepsilon_L + \varepsilon_S = 1$ 7.3

The overall gas holdup was evaluated from the expanded bed height as in the case of air-water and systems containing single and binary components particle using equation 5.1 (i.e., $\varepsilon_g = (Hf - H_O)/Hf$).

A comparison and discussion of the results is given in chapter 10.

7.3 EXPERIMENTAL RESULTS

Tables of the experimental results are provided in appendix D.

7.3.1 GAS HOLDUP

The trends obtained were similar to those observed for three phase systems containing single component and a binary mixtures of particles. However , further decreasing, but very small, effect on gas holdup was produced by the presence ternary mixture

EFFECT OF TERNARY MIXTURE CONCENTRATION AND CONCENTRATION RATIO.

(1) TOTAL CONCENTRATION

Two total binary mixture concentration levels were used - 10% and 20% packed volume/volume, of the total system. Table D1 in appendix D gives the results for ternary mixture MT1 (Ballotini/Styrocell/Amberlite) of equal size and concentration ratio.

On increasing the total concentration of the ternary particle mixture from 10% to 20%, the reducing effect caused by the presence of the additional solid compoment was diminished, particularly at high gas velocities, and results were close to those obtained for air-water-(Ballotini/Styrocell binary mixture).

(2) <u>CONCENTRATION RATIO</u>

Table D2 in appendix D shows the effect of concentration ratio for all the ternary mixtures used. Again, as suggested in chapter 6, individual particle components appeared to be in competition, demonstrating yet again a strong "trade-off" of properties. Generally , the combination of Ballotini and Styrocell produce a clear reduction in gas hold-up at high superficial gas velocities.

EFFECT OF SIZE AND SIZE RATIO

For ternary mixtures MT1 and MT3 , Ballotini and Styrocell were in reversed size ratio. From Table D3 in appendix D, marginally higher holdups (up to 2.5% up) were recorded for MT1 (compare table D3 and table C3) . There were no difference between MT3 and M1b, size ratio 2:1.

EFFECT OF DENSITY AND DENSITY RATIO

The results are given in Table D4 in appendix D, for ternary mixtures MT1 and MT3. Gas holdup values for MT1 (Balotini/Styrocell/Amberlite) agrees reasonally well with MM1a (Ballotini/Styrocell) binary mixture.But the results are higher for systems MT3, Ballotini/Styrocell?Diakon.

7.3.2 SOLID HOLDUP

The overall solids holdup reduced with increasing superficial gas velocity at any axial position irrespective of the ternary mixture concentration and size and density ratios .

EFFECT OF CONCENTRATION AND CONCENTRATION RATIO

Table D5 in appendix D gives the results obtained for all ternary mixtures. Increasing the total concentration of the mixture only produces shifts in the holdup variation similar to those obtained from the systems containing binary particles. However, the overall holdup contribution from each solid in the ternary system was dependent on the concentration ratio, amongst other parameters.

EFFECT OF SIZE AND SIZE RATIO

Table D6 in appendix D shows the effect of size and size ratio on solid holdup of MT1 and MT2. Results presented in the table were typical for MT3.

EFFECT OF DENSITY AND DENSITY RATIO

The solid holdup is found to be sensitive to density , but not to density ratio. . Tables of experimental values are listed in Table D6 and D7 in the appendix D show very little difference.

CHAPTER EIGHT

FLOW VISUALISATION STUDY

8.1 INTRODUCTION

The flow regimes and liquid circulation patterns that existed under the experimental conditions of the present work are presented in this chapter. Visual observations were in good agreement with the generally accepted flow characteristics described in the literature.

The results are presented in a format based on the effects of operating parameters: solid size, concentration, density and nature of the solids. The effect of the ratio of these parameters is assessed in the case of systems containing binary particle mixtures. A qualitative analysis of mixing and the flow patterns in three-phase fluidised beds containing the binary mixtures is also given.

The characteristics of a flow regime (section 2.4) depend on various parameters. Of these characteristics, one of the most important is the size of the bubbles; this parameter along with the gas holdup determines the surface area available for mass and heat transfer in gas- liquid and gas-liquid-solid fluidised operations. Of equal importance are the bubble rise velocity, bubble size distribution and liquid and bubble velocity profiles: all have direct bearing on the problems associated with the design of bubble column reactors.

Liquid circulation is a prominent feature of flow in the churn-tubulent regime in a bubble column. Large bubbles and slugs are driven towards the central region, and large liquid eddies (circulation cells) in orderly patterns can frequently be observed. In the most common type of circulation, called "gulf-streaming", liquid carrying a high concentration of gas bubbles rises upward near the center of the column; liquid carrying fewer bubbles flows back downward near the walls of the column. The central gulf-stream serves to carry gas through the column more rapidly than is the case when it is transported in the form of bubbles evenly distributed across the column diameter (ie in the bubbly flow regime) and as a result, the efficiency of the column as a reactor is altered.

Solid mixing in a gas-liquid-solid fluidised system is characterised by axial mixing, wake phase and bulk phase exchange, segregation and bulk circulation; all are influenced by bubble dynamics. In three-phase systems, the gas flow dominates the liquid mixing and circulation and this in turn influences particle movement. The consequence is that segregation of the solid components is suppressed.

8.2 EXPERIMENTAL PROCEDURE

Experimental investigations were carried out in the two-dimensional bubble column, already described in chapter 4. The bubble column was well lit at the back. Air and water were employed as the dispersed gas and fluidising liquid respectively. Ballotini, Styrocell, Amberlite, and Diakon were used to examine the effect of varying the properties of the solid phase.

The experimental procedure was as described in chapter 4. Still photographs of the column were taken, using a canon AEI programable camera. A JVC colour video camera was also used to provide a permanent record of each experiment. This allowed adequate checks to be made of the visual observations recorded during each experiment in terms of bubble coalescence and break-up, liquid circulation and solid phase mixing and segregation. Exact bubble, eddy and circulation cell sizes could not be obtained by this method, but acceptable estimates were possible. The experimental set up is illustrated in figure 8.1.

8.3 EXPERIMENTAL RESULTS

8.3.1 THE AIR-WATER SYSTEM

Because of the availability of adequate information on flow regimes and bubble dynamics in two-phase, air-water systems in the literature (see chapter 3), this system was employed for reference purposes when interpreting visual observations made with the more complex three-phase systems. In particular, the classification proposed by Wallis (1969) for upward movement of bubbles swarms in a bubble column was used (see chapter 2).

Figure 8.2 shows the photographic results for the air-water system at diferent superficial gas velocities and zero superficial liquid velocity. At a superficial gas velocity, Usg = 3.10cm/s (figure8.2a), there was a relatively homogeneous distribution of small bubbles of similar sizes (typically 3-5 mm). It will also be observed that there is evidence of bubble coalescence and cluster formation at the centre of the column. These features have previously been reported by Fair (1967). It should be noted that similar behaviour was observed in the top and bottom sections of the column.

At higher gas velocities, a homogeneous gas dispersion could not be sustained and the flow patterns became unstable. Large bubbles moving at high velocity within the dispersion of small bubbles were observed. Rapid coalescence of bubbles was a

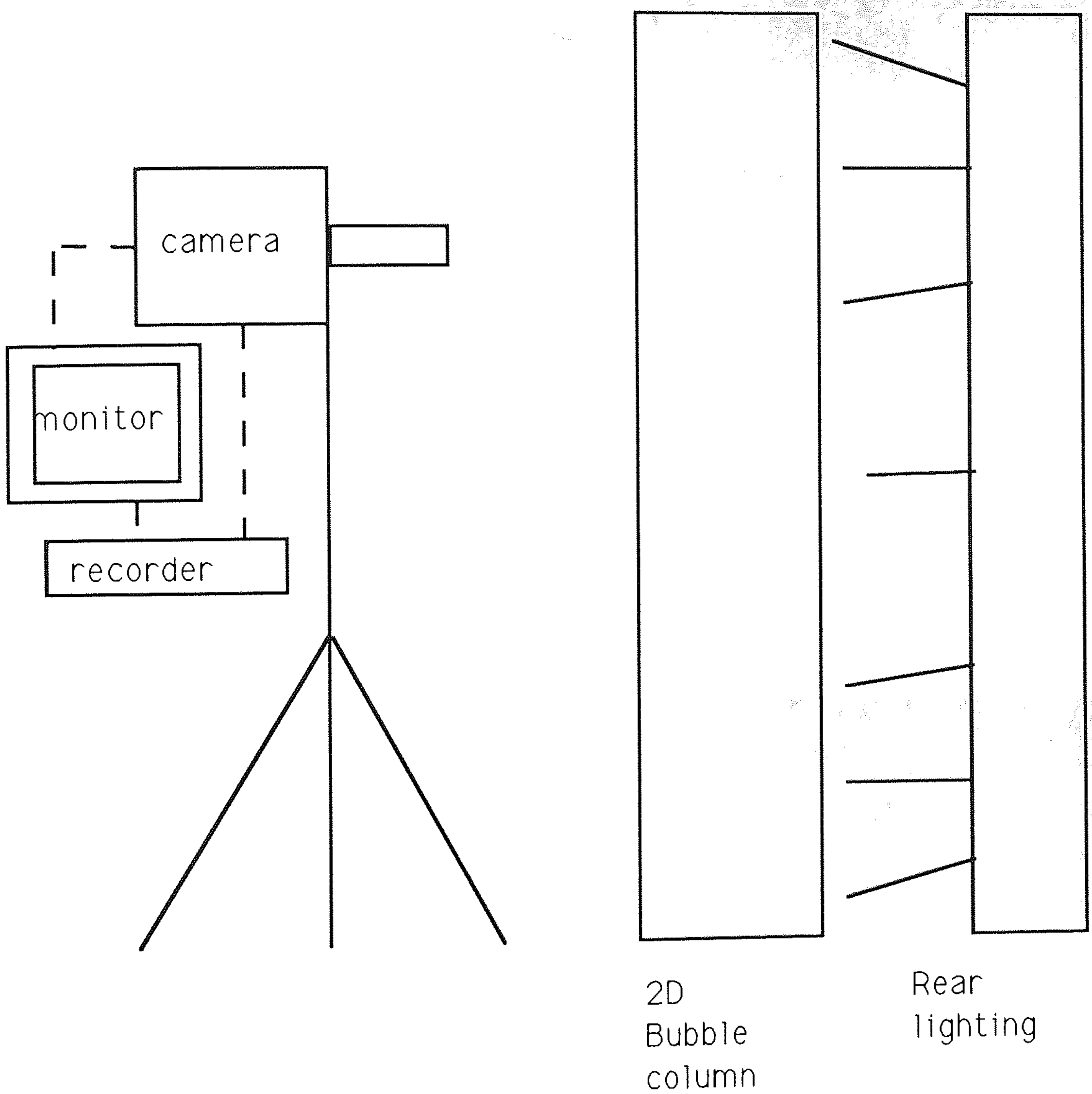
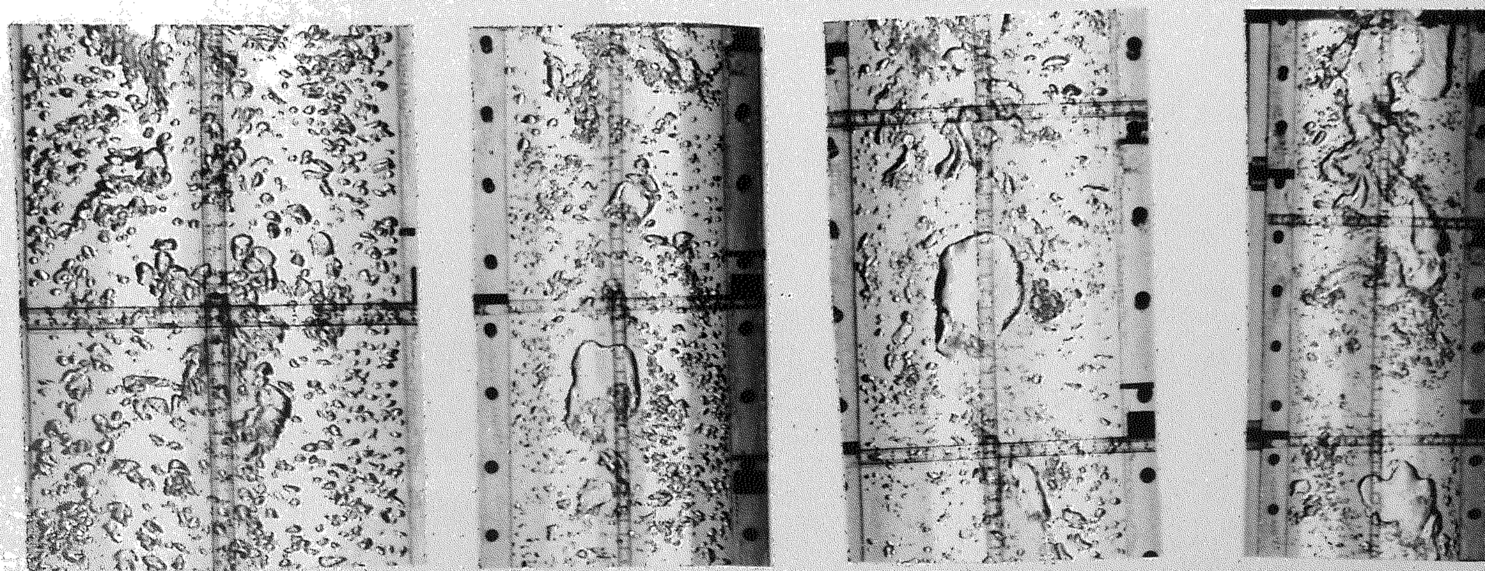


Figure 8.1: A two-dimensional representation of experimental setup for visualisation studies.

Figure 8.2 : Flow Regime in two-phase (Air-Water) fluidised bubble column

(a) (b) (c) (d)



	(a)	(b)	(c)	(d)
Usl=0				
Usg (cm/s)	3.10	11.45	15.23	19.21

particular feature of this regime, particularly near the top of the column and at the upper
surface (see figure 8.2 b and c). As the superficial gas velocity was further increased, elongated, flattened slugs, centrally positioned dominated the scene. Under these conditions bubble breakup and coalescence was violent, generating smaller bubbles, (see figure 8.2d).

8.3.2 AIR-WATER-SINGLE COMPONENT SOLIDS

EFFECT OF OPERATING PARAMETERS

Figures 8.3 to 8.5 illustrate the effect of operating parameters on the behaviour of air-water systems containing each of the four solids used in this study. Bubbles / slug sizes increased with increasing superficial gas velocity in all cases. The bubbly regime (Usg < 3-5 cm/s) observed with air-water systems was entirely absent. The occurence of bubble swarms appeared to have been suppressed by the presence of solids, and large discrete bubbles with movement orientated centrally were observed. In the case of air-water-Ballotini, the bubbles were approximately 2cm in size as they emerged from the bed surface and increased up to approximately 4.5 cm at the top of the column, (figure 8.3). The author attributes this to the fact that, for air-water-ballotini systems, in which the bottom section behaves as a packed bed, mal-distribution of gas occurs. Above a superficial gas velocity, of 3-5cm/s the flow regimes observed were more similar to those observed for the air-water system. However important variations were still apparent due to other factors, discussed below.

In general, the rate of bubble coalescence and bubble break-up was also a strong function of superficial gas velocity. Coalescense occured most noticeably about the middle of the column, while bubble breakup took place more readily near the top and at the interface at the top of the column.

The amount of solids mixing within the liquid phase was a strong function of superficial gas velocity, particularly for air-water-Ballotini. For other solids with densities nearer to that of water, the mixing time was a stronger function of gas velocity.

EFFECT OF SOLIDS CONCENTRATION

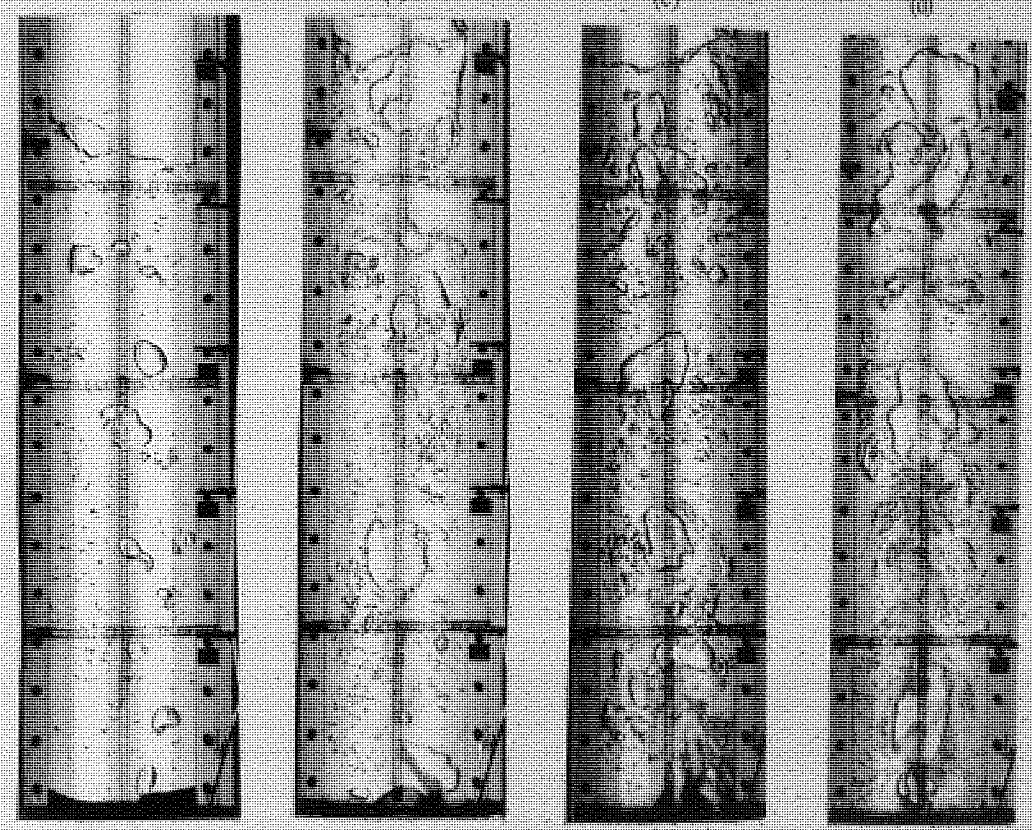
Figure 8.4 shows the typical effects produced by increasing solids concentration from 10% to 20% volume by volume of the total system before fluidisation, for air-water-

Figure 8.3 : Effect of superficial gas velocity (air-water-ballotini)

(i) 600μm particles ;

10% v/v

(a) (b) (c) (d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

(ii) 600μm particles;

20% v/v

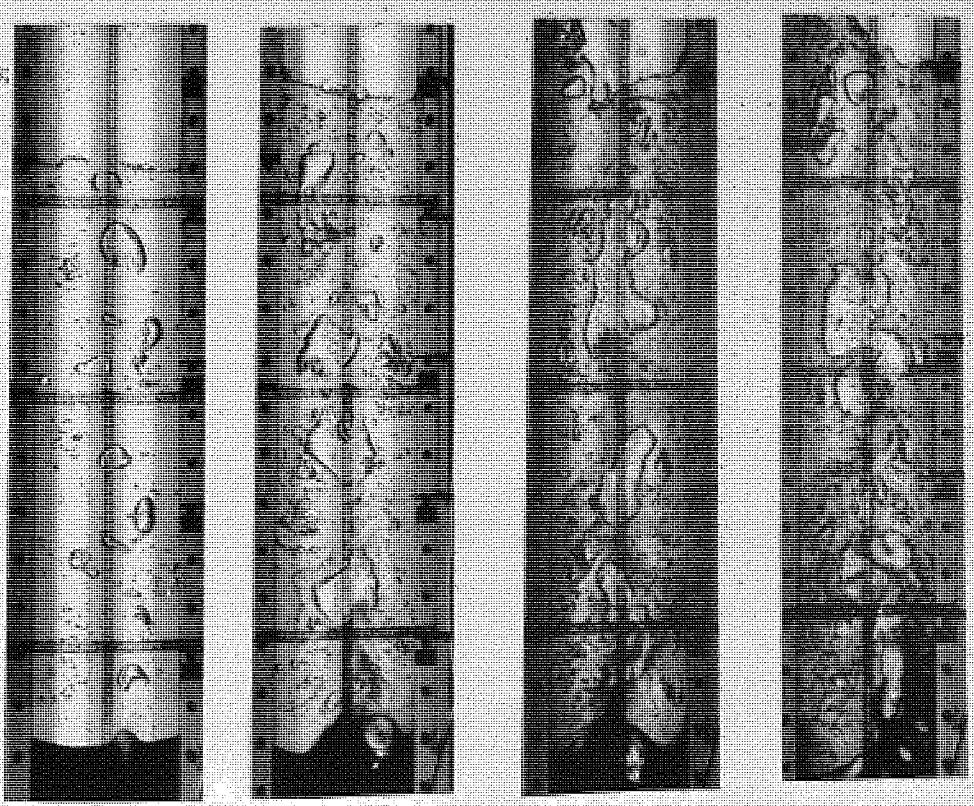


figure 8. 3 contd.

(iii) 1200μm particle;
20% v/v

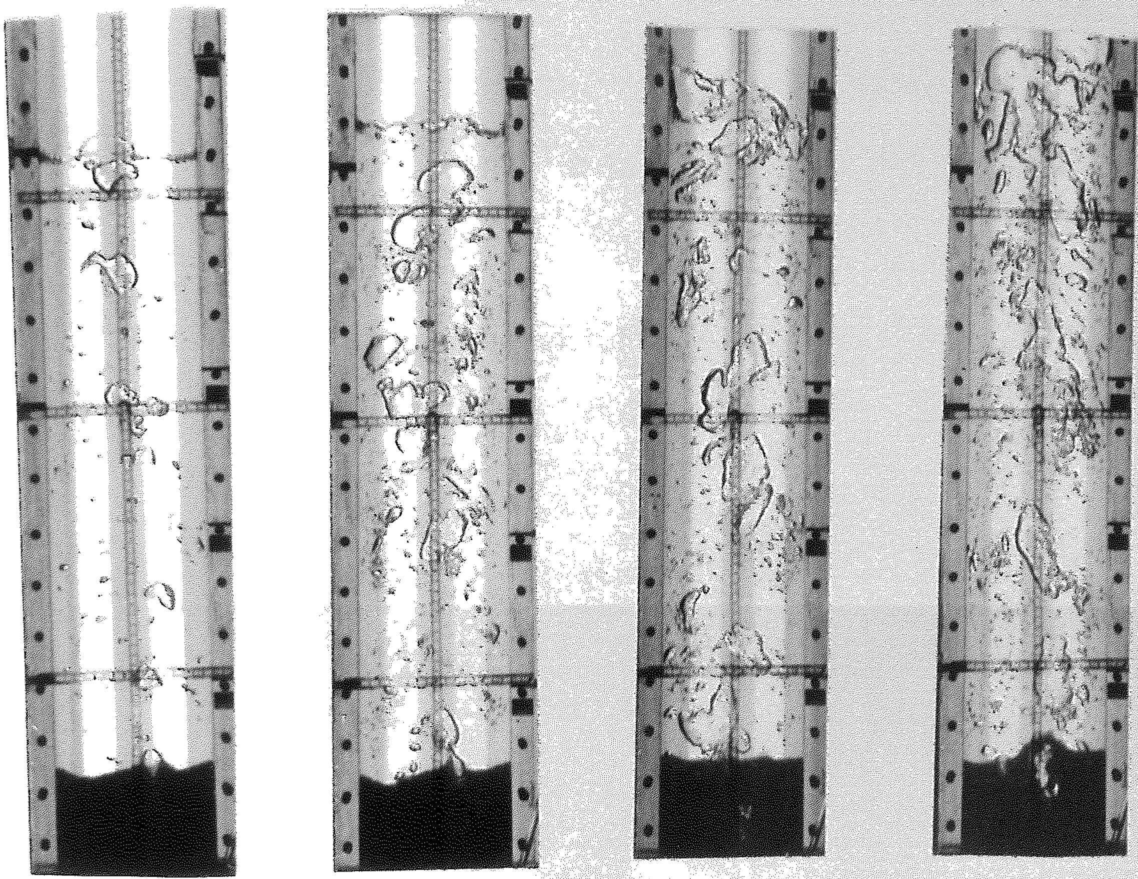


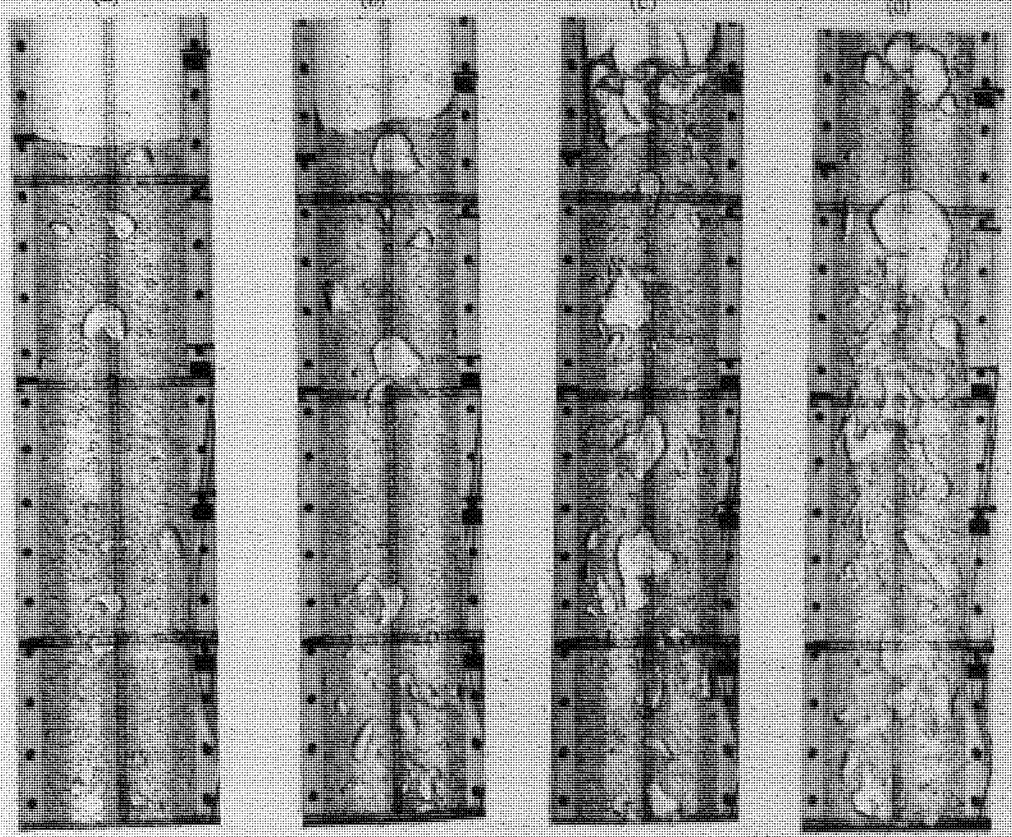
Figure 8.4 : Effect of solid concentration and size

air-water-styrocell

(i) 1200μm particles ;

10% v/v

(a) (b) (c) (d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

(ii) 1200μm particles;

20% v/v

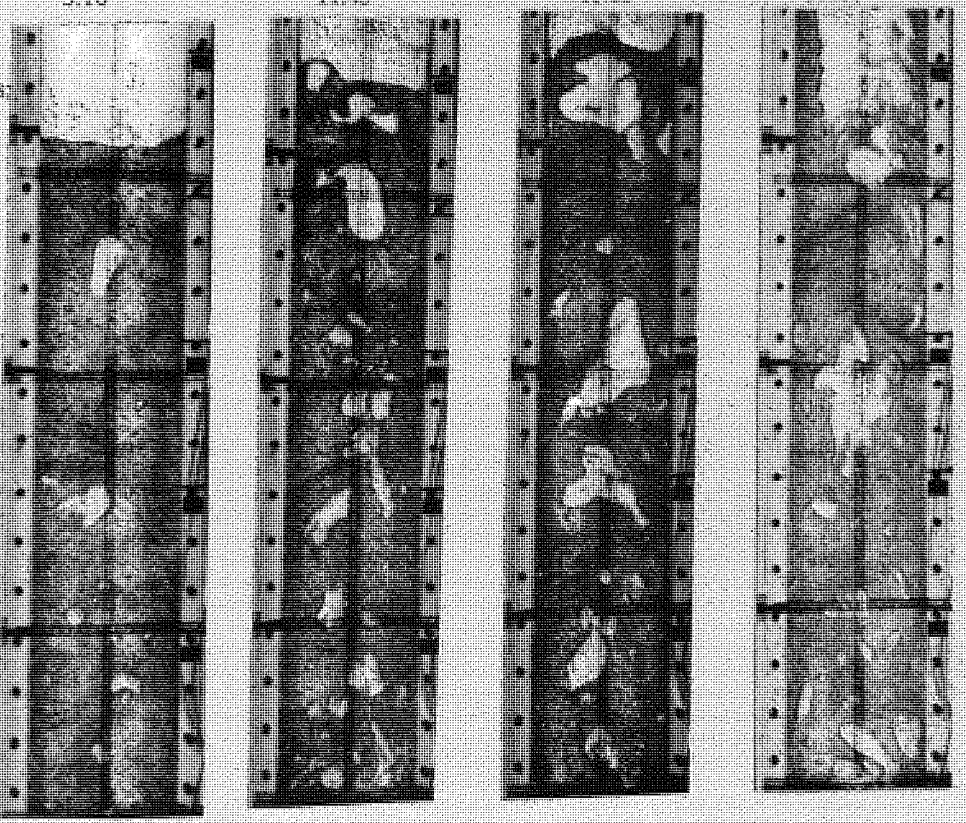
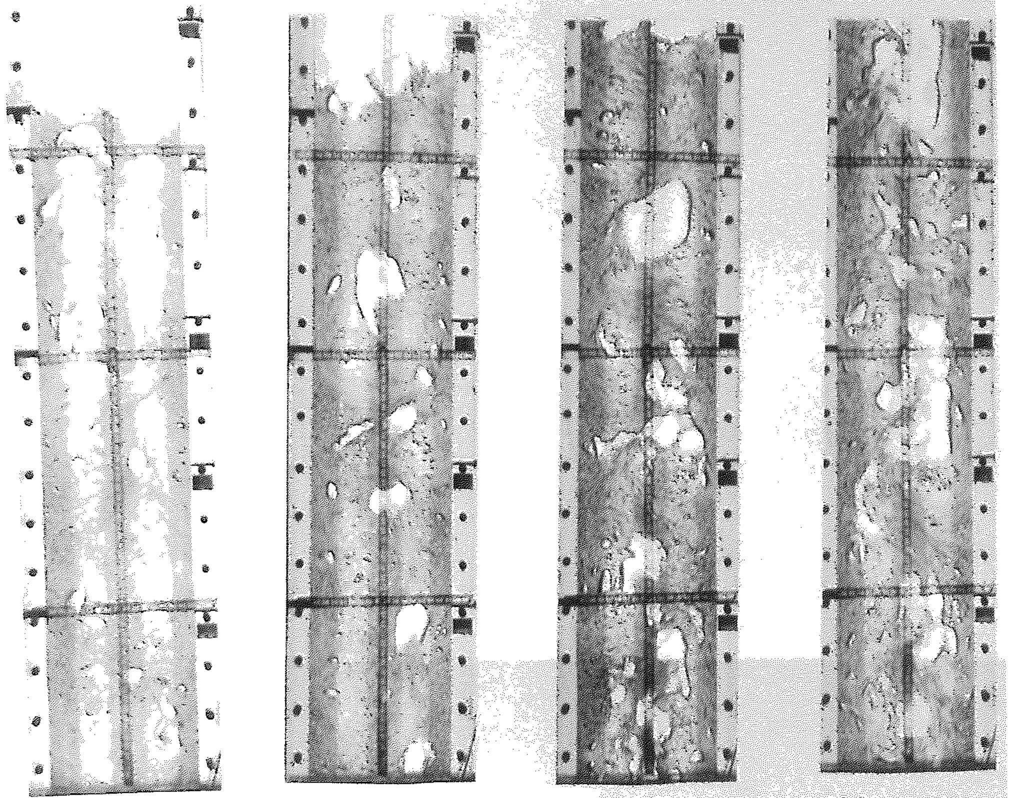


figure 8. 4 contd.

air-water-amberlite

(iii) 600μm particles ;

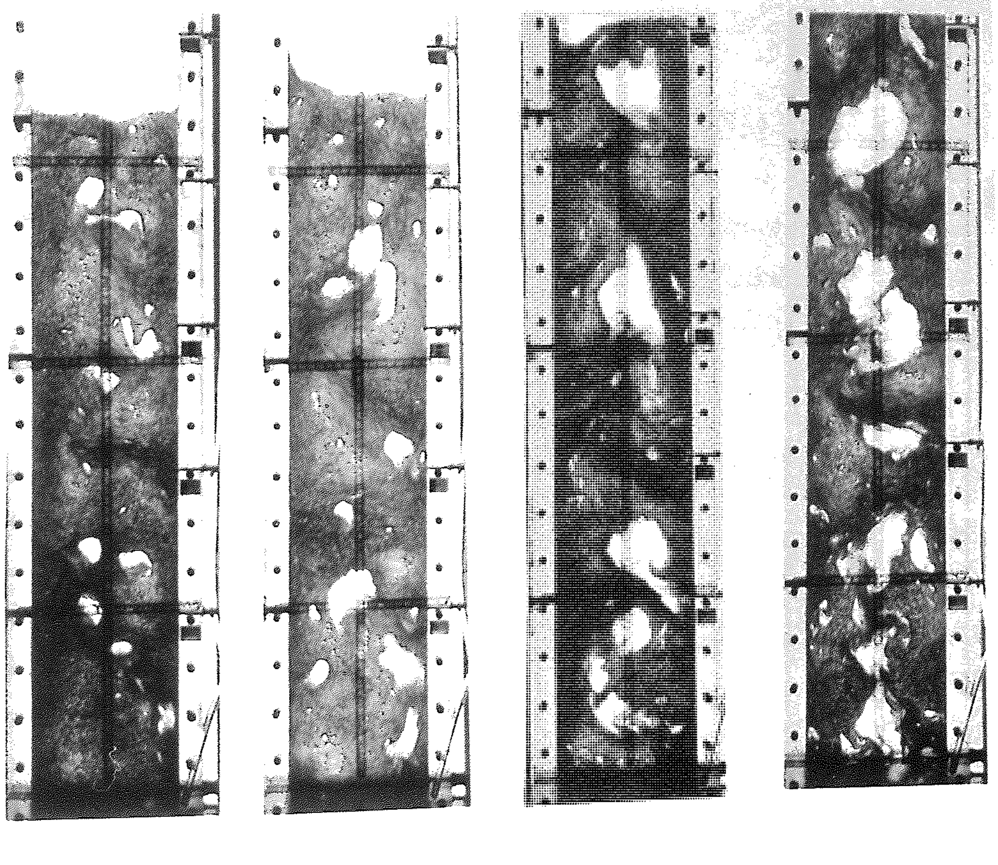
10% v/v



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

(iv) 600μm particles;

20% v/v



Styrocell and air-water-Amberlite. Deviations from bubbly flow were observed at all gas velocities much less than 3-5cm/s. Increasing the solids concentration also delayed the point at which the fully developed churn-turbulent state was attained. Fewer bubbles were present over the entire gas velocity range; also, bubble coalescence was reduced in the presence of the Amberlite resin. Because mixing of these solids was relatively easy, the axial concentration was constant.

EFFECT OF PARTICLE SIZE

Generally, no significant effect of particle size was apparent. However the development of the churn-turbulent regime was somewhat delayed in the case of the air-water-Ballotini system when using particles 1200μm in size rather than those 600μm in size.

EFFECT OF SOLID DENSITY

The effect of solid density on the flow regime was significant when the behaviour of systems containing ballotini (ρ_p - 2.84 to 2.92 g/cm^3) was compared with that of other systems (ρ_p - 1.11 to 1.28 g/cm^3) at superficial gas velocities less than 3 - 5 cm/s. However, when the results for air-water-Diakon were closely examined over this gas velocity range, the gradual transition to full churn-turbulent flow could be due to parameters other than solid density. Above this velocity, no significant effect of density was noticed. Solid density does, of course, play an important role in the fluidisability of the solid phase.

EFFECT OF THE NATURE OF THE SOLID PHASE

Previous reseachers at Aston have classified the four solids used in these experiments into two groups based on their "wettability". The author has not found this classification to be helpful because of the significant differences observed for all the systems: also, there was overlap in the observations with Amberlite and Diakon as well as with the others. Therefore, the effect of the surface property is somewhat unclear.

AIR-WATER-DIAKON

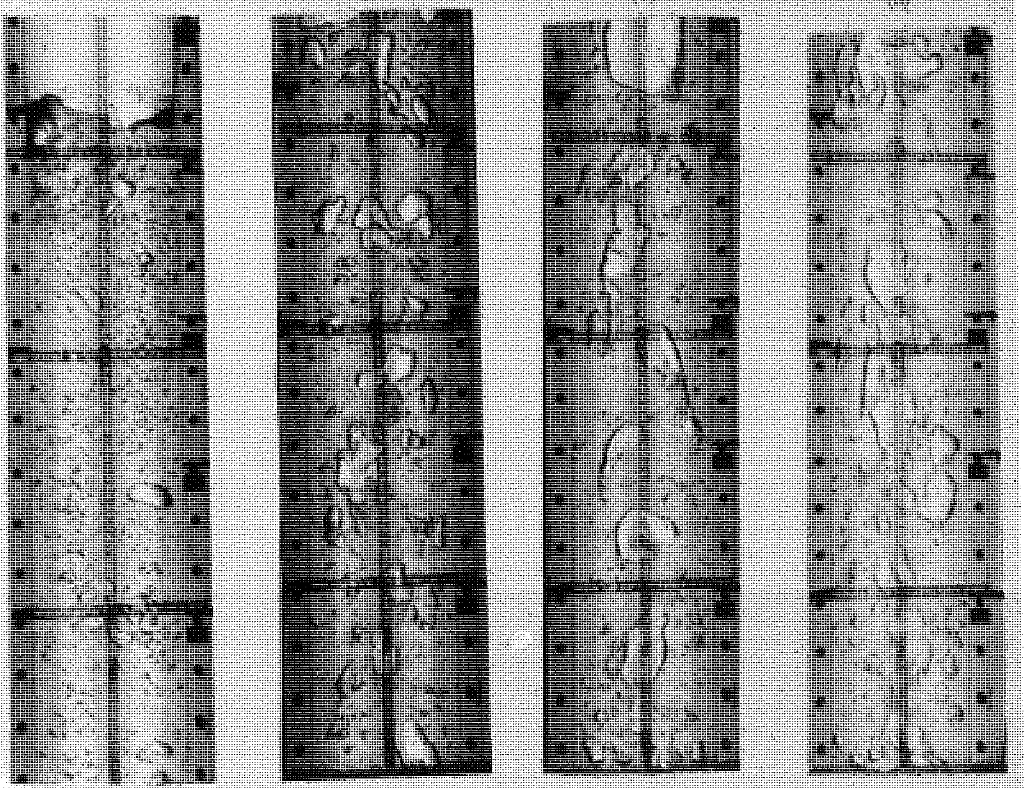
Figure 8.5 shows typical photographs of the air-water-Diakon system. Bubble coalescence was noticeably reduced with this system and also with the air-water-Amberlite system. A regime similar to homogeneous bubbly flow was observed at gas velocities < 5cm/s. This feature was due to regeneration of identical smaller bubbles

Figure 8.5: Effect of wettability property of solids

air-water-diakon

(i) 600μm particles ;
10% v/v

(a) (b) (c) (d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

(ii) 600μm particles;
20% v/v

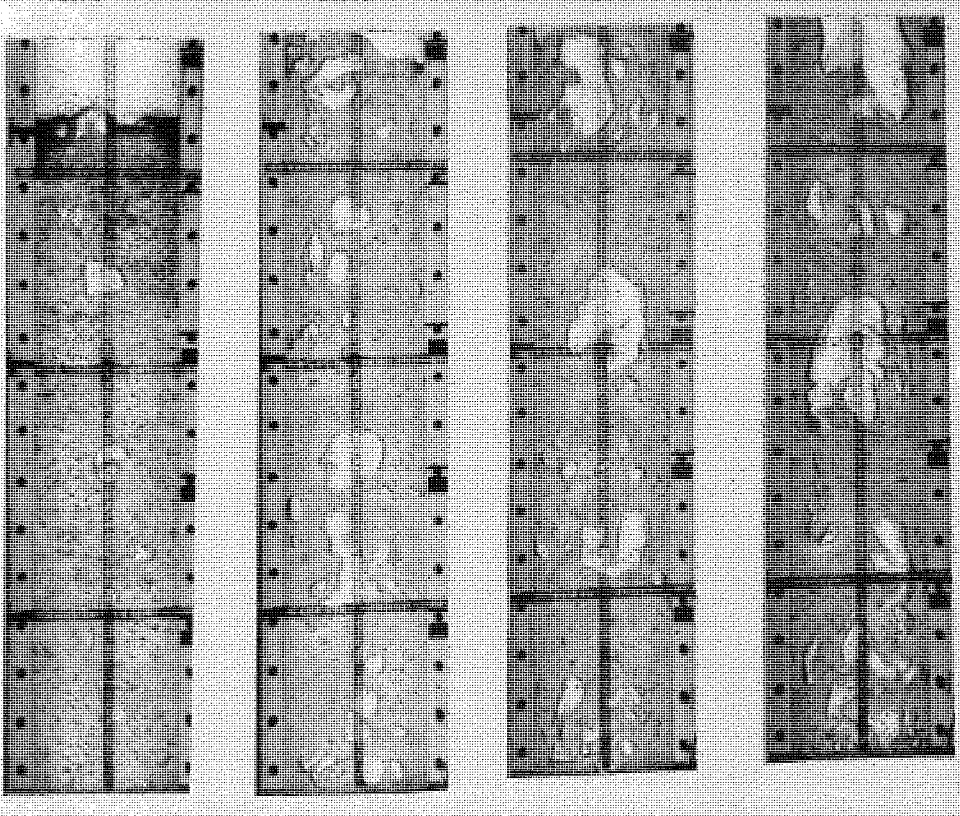


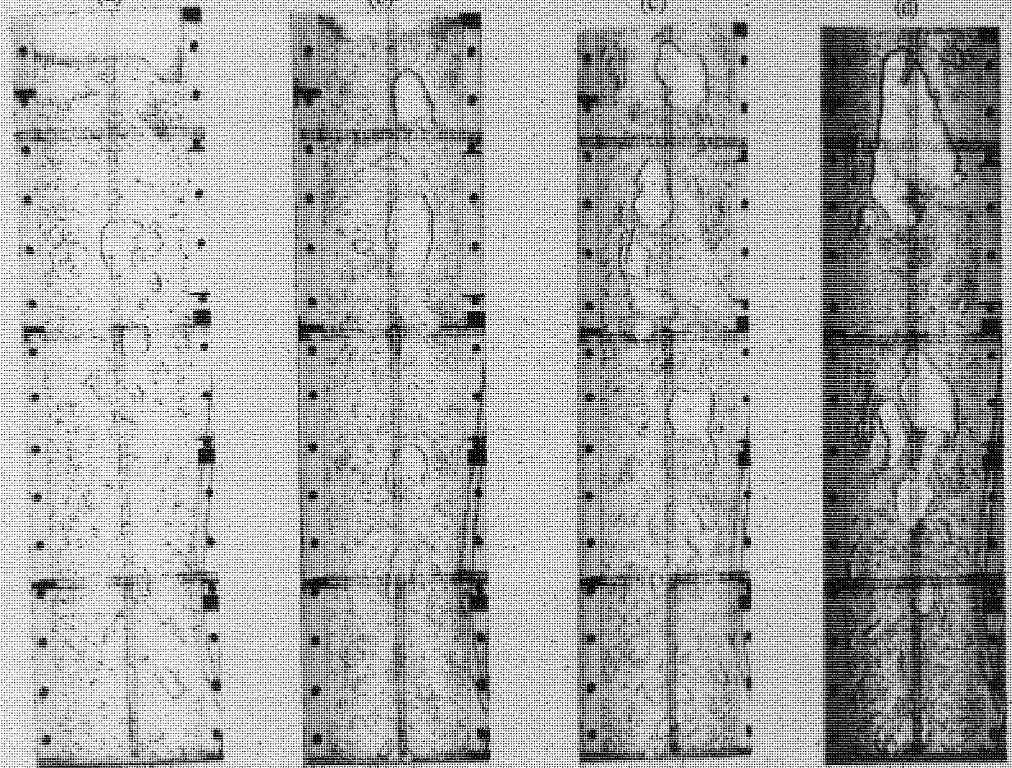
figure 8. 5 contd.

air-water-diakon

(iii) 1200μm particles ;

10% v/v

(a) (b) (c) (d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

(iv) 1200μm particles;

20% v/v

after bubble breakup at the top of the column. These "secondary" bubbles were recirculated throughout the system, an observation which was not made for Amberlite. Strong adhesion of the solids to the bubble envelope was a significant characteristic of this system; and occurred more readily with the 600µm particles and on increasing the solids concentration. Some solid adhesion to the bubbles was evident in the case of the Styrocell particles and amberlite resins; this was short-lived and limited to low gas velocities. Thus, there appears to be a "trade-off" or interraction between solids density, the "wettability" , concentration and size properties of the solids.

8.3.3 AIR-WATER-BINARY MIXTURES OF SOLIDS

EFFECTS OF OPERATING PARAMETERS

Similar observations were made as with systems with single component solids on increasing superficial gas velocity. However, large bubbles due to bubble coalescence were observed with all systems used even at gas velocities < 5cm/s. Figures 8.6 to 8.8 show the flow regimes observed for each system. Solids mixing was a strong function of superficial gas velocity.

EFFECT OF SIZE RATIO

Figure 8.6 shows the results obtained for systems containing three different binary mixtures of Ballotini / Styrocell, at 1:1, 1:2 and 2:1 size ratio at equal amount (all binary mixture ratios are shown in table 6.1), and two total particle concentration levels, 10% and 20% volume by volume. The results are typical of those made with other systems containing binary mixtures except for systems containing Ballotini / Diakon (see figure 8.8), which showed some differences. This system appeared to divide into three sections: a packed section at the bottom containing essentially ballotini, whatever the size ratio, followed by a thin clear region (<1cm in depth), and a fluidised top section which contained essentially the lighter particles. Hence, the flow regimes were identical to those recorded for the single, lighter particles. When the size ratio was reversed (see figures 8.6b and c), this did not appear to have any marked effect on the observed flow regimes. However, bubble coalescence and bubble breakup were enhanced.

EFFECT OF CONCENTRATION RATIO

Figure 8.7 provides the photographic evidence of the effect of concentration ratio for Styrocell / Amberlite binary particle mixtures. Both solid components were fully

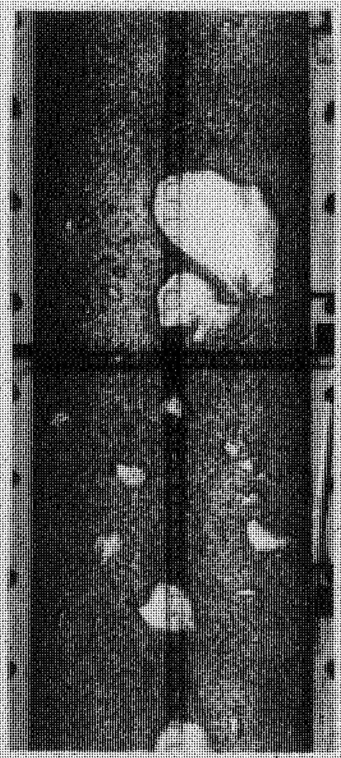
Figure 8.6: Effect of particle size ratio on a three-phase system containing binary particle mixture ballotini \ styrocell

(i) size and mixture ratio 1:1

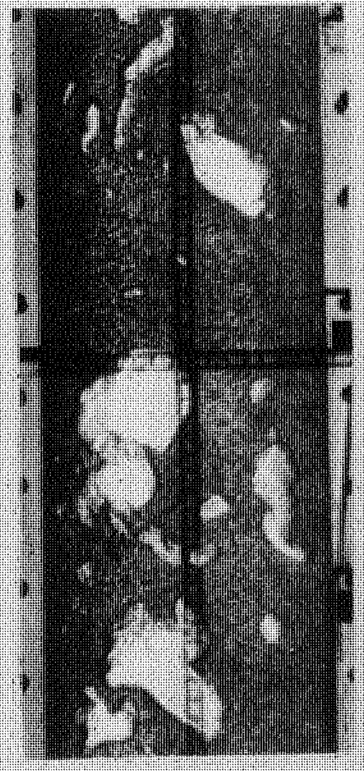
total mixture 20% v/v

(a)

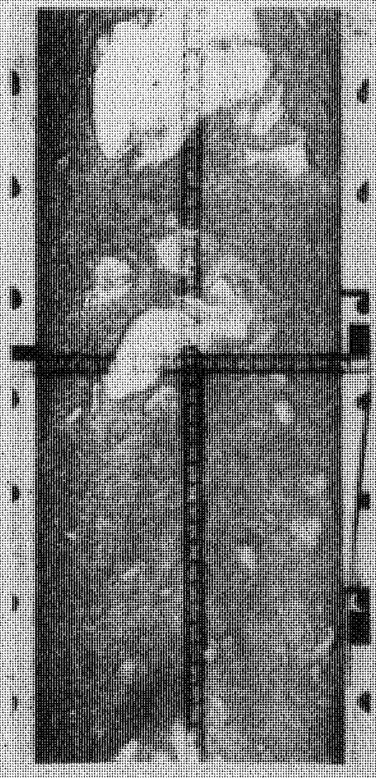
(b)



(c)



(d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

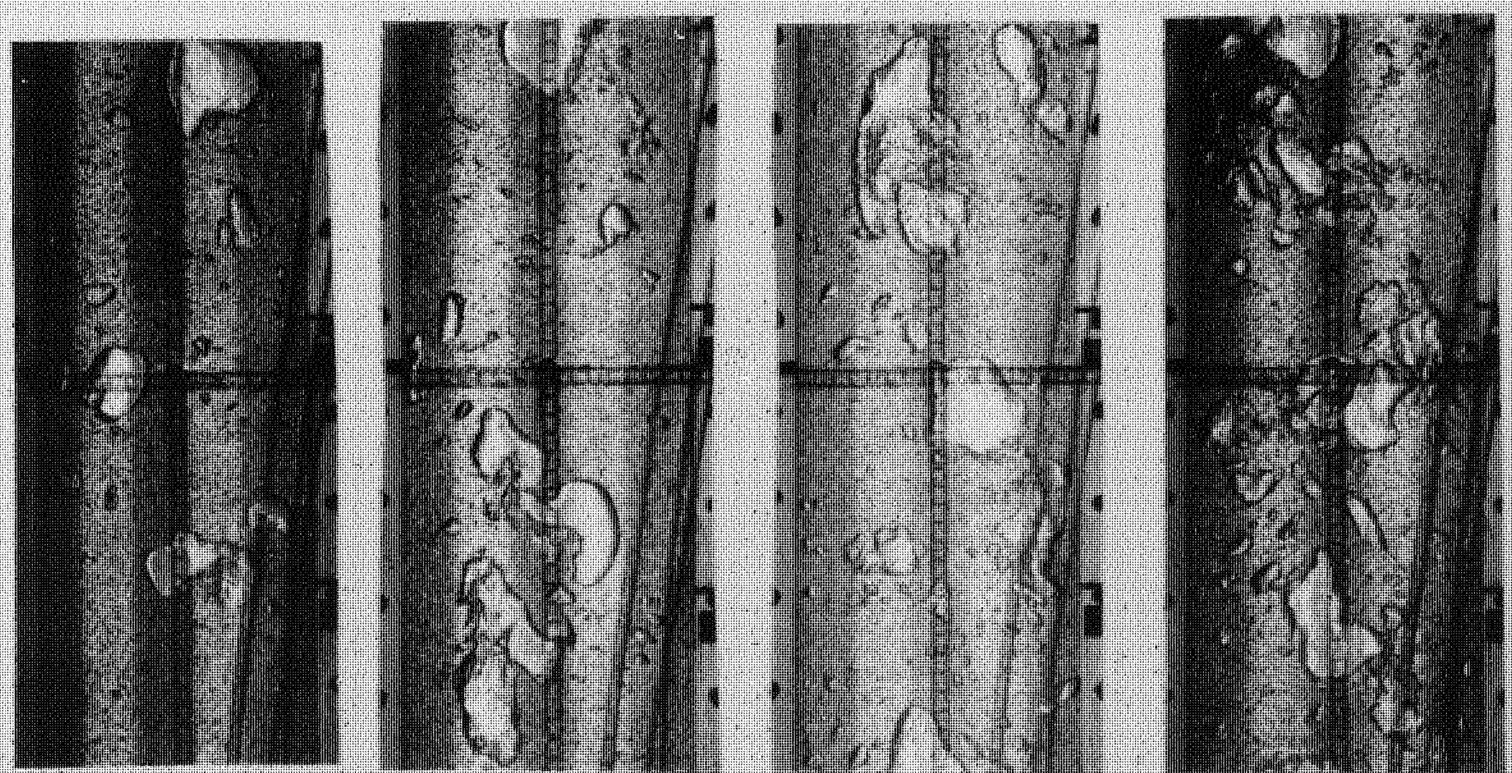
Figure 8.6 contd.

(ii) size ratio 1:2

mixture ratio 1:1

total concentration 20%

(a) (b) (c) (d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

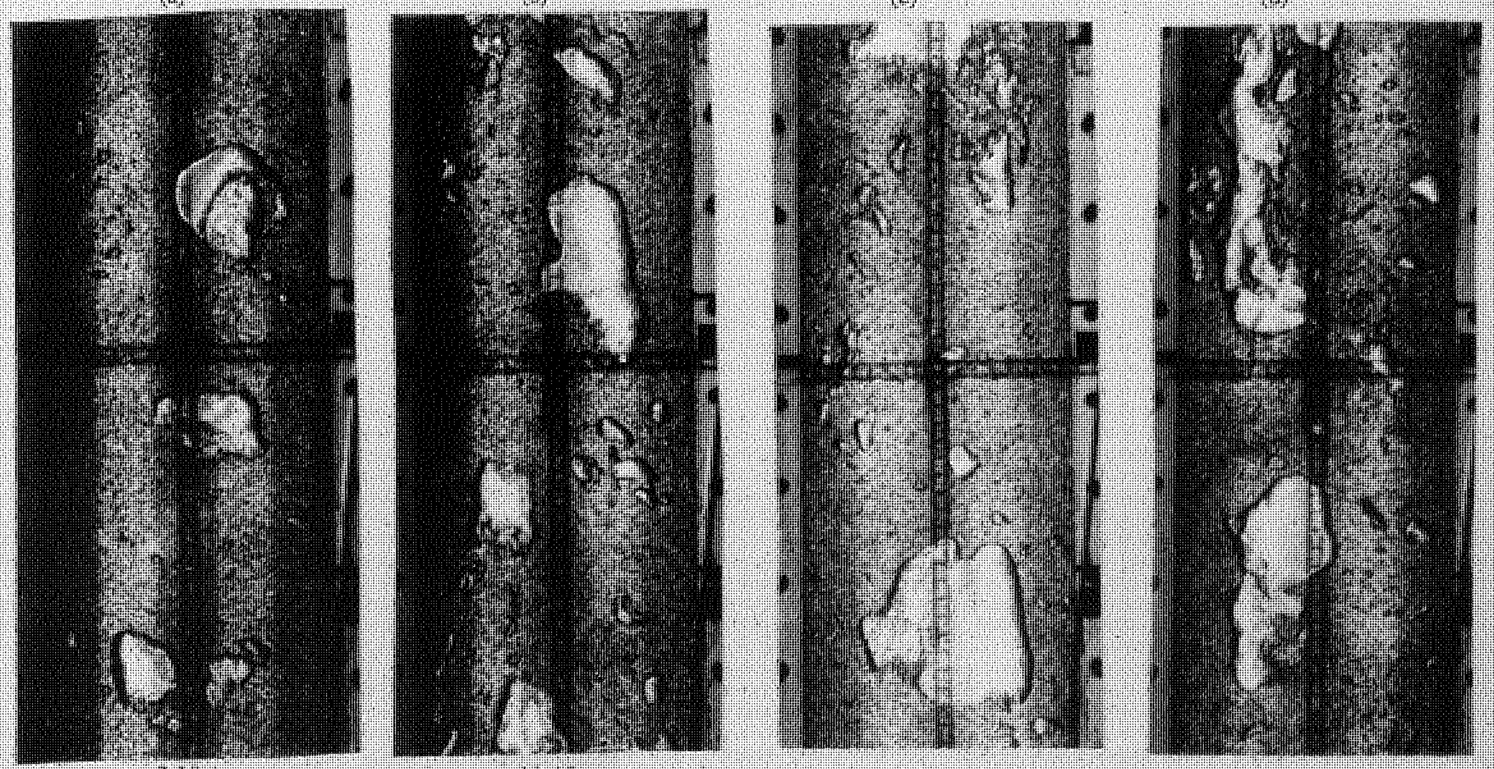
Figure 8.6 contd.

(iii) size ratio 2:1

mixture ratio 1:1

total mixture 10%

(a) (b) (c) (d)



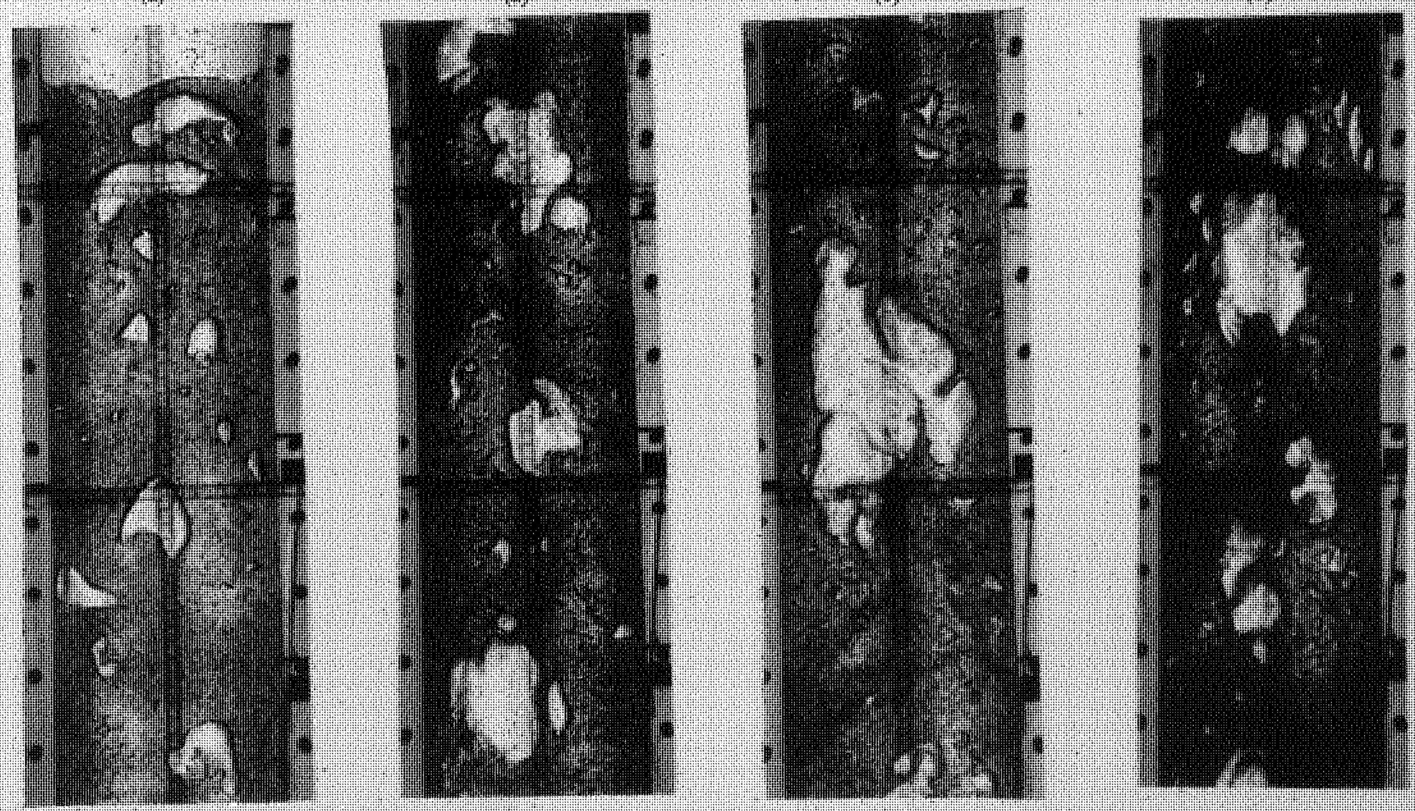
U_{sg} (cm/s) 3.10 11.45 15.23 19.21

Figure 8.7: Effect of solid concentration ratio on three-phase system containing binary particle mixture
(size ratio 1: 2; total solids 20% v\w)

styrocell \ amberlite particle mixture

(i) concentration ratio 1: 2

(a) (b) (c) (d)

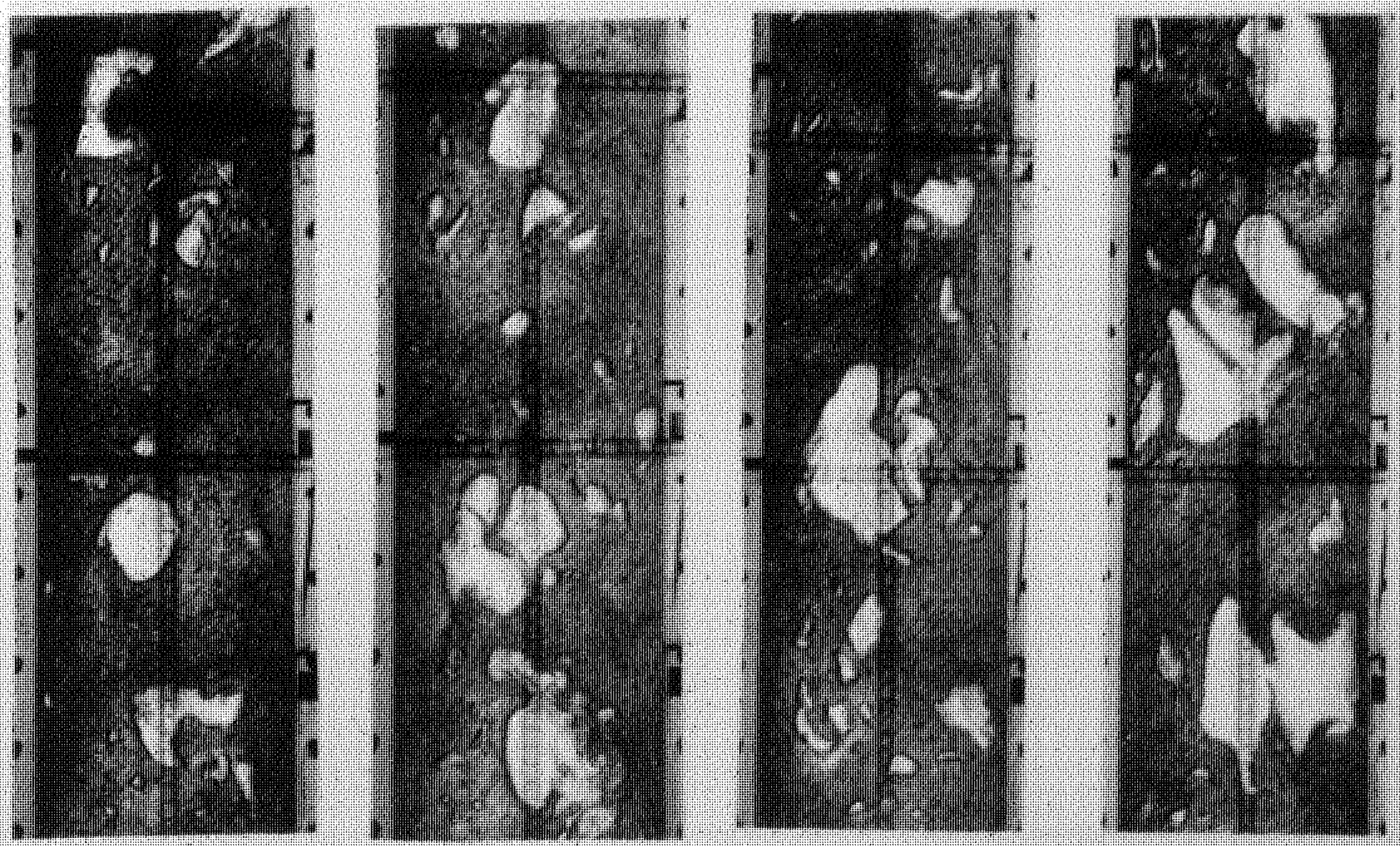


U_{sg} (cm/s) 3.10 11.45 15.23 19.21

Figure 8.7 contd

(ii) concentration ratio 2: 1

(a) (b) (c) (d)



U_{sg} (cm/s) 3.30 11.45 15.23 19.21

suspended in the three-phase system. Coalescence of bubbles was encouraged to give flattened, large bubbles over the entire range of superficial gas velocity.

EFFECT OF DENSITY RATIO

Figure 8.8 provides a comparison of the results obtained using three binary mixtures: Amberlite / Diakon (density ratio 1.06), Ballotini / Diakon (density ratio 2.21) and Ballotini / Diakon (density ratio 2.63), all at equal concentration.

Bubble dynamics showed a significant variation at high gas velocities (ie > 5cm/s). Bubble coalescence was promoted by increasing the density ratio: however, this particular parameter was not thought to provide the only reason for this findings. The high degree of interraction between other properties will contribute to the effect found.

EFFECT OF THE NATURE OF SOLID MIXTURE

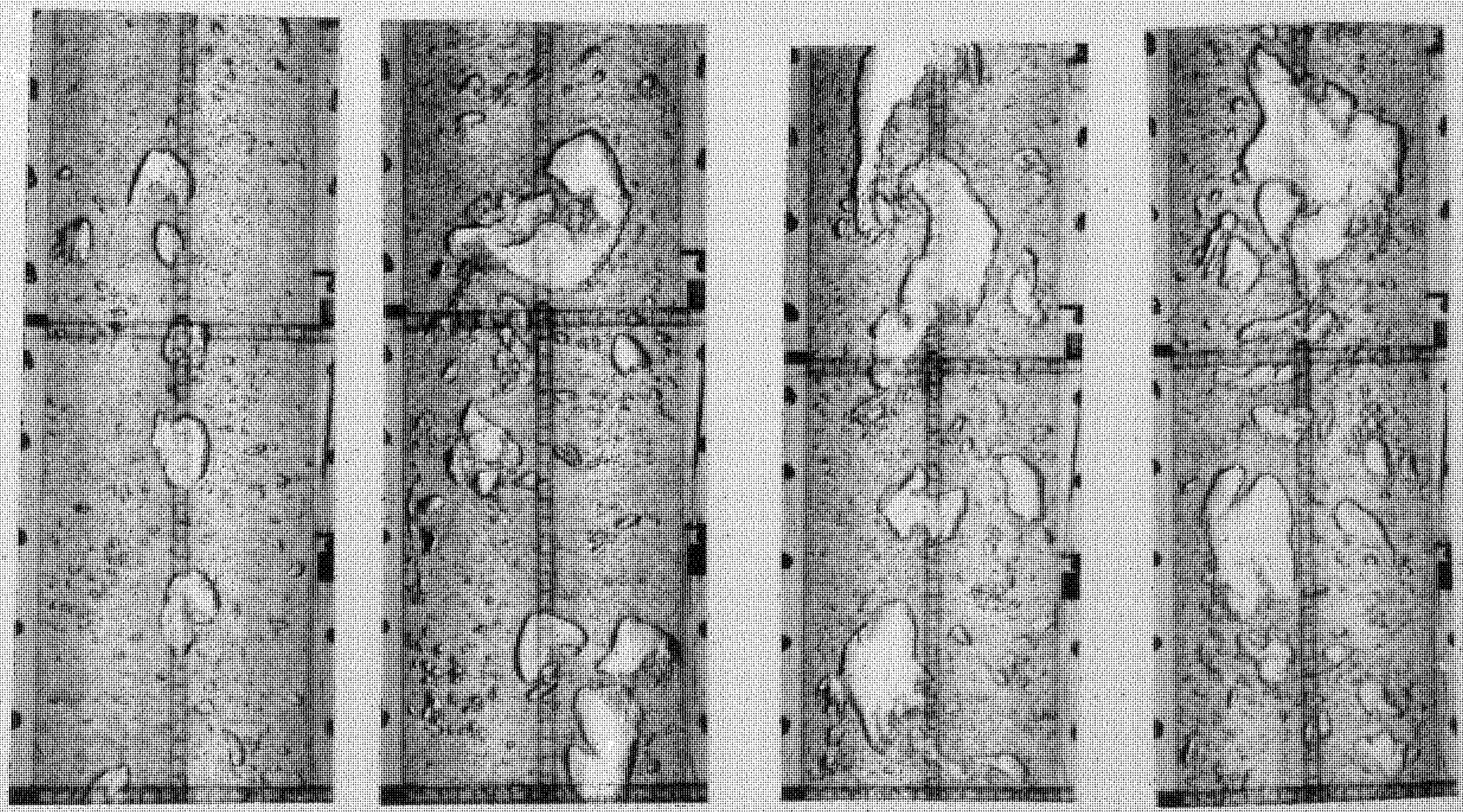
The significance of the nature of the solid particles only showed up in mixtures containing Diakon as one of the components. The flow characteristics observed were similar to those recorded for systems containing Ballotini only (cf. figure 8.3 and 8.8). All the Diakon particles were suspended, while only about one-third of the ballotini particles were fluidised with the remainder forming a packed bed. The significant generation of smaller bubbles over the gas velocity range was in agreement with observations made for systems containing Diakon only (figure 8.5).

Figure 8.8: Effect of density ratio on three-phase system containing binary particle mixture

amberlite\diakon

(i) density ratio 1.06

(a) (b) (c) (d)



U_{sg} (cm/s)	3.10	11.45	15.23	19.21

Figure 8.8 contd.

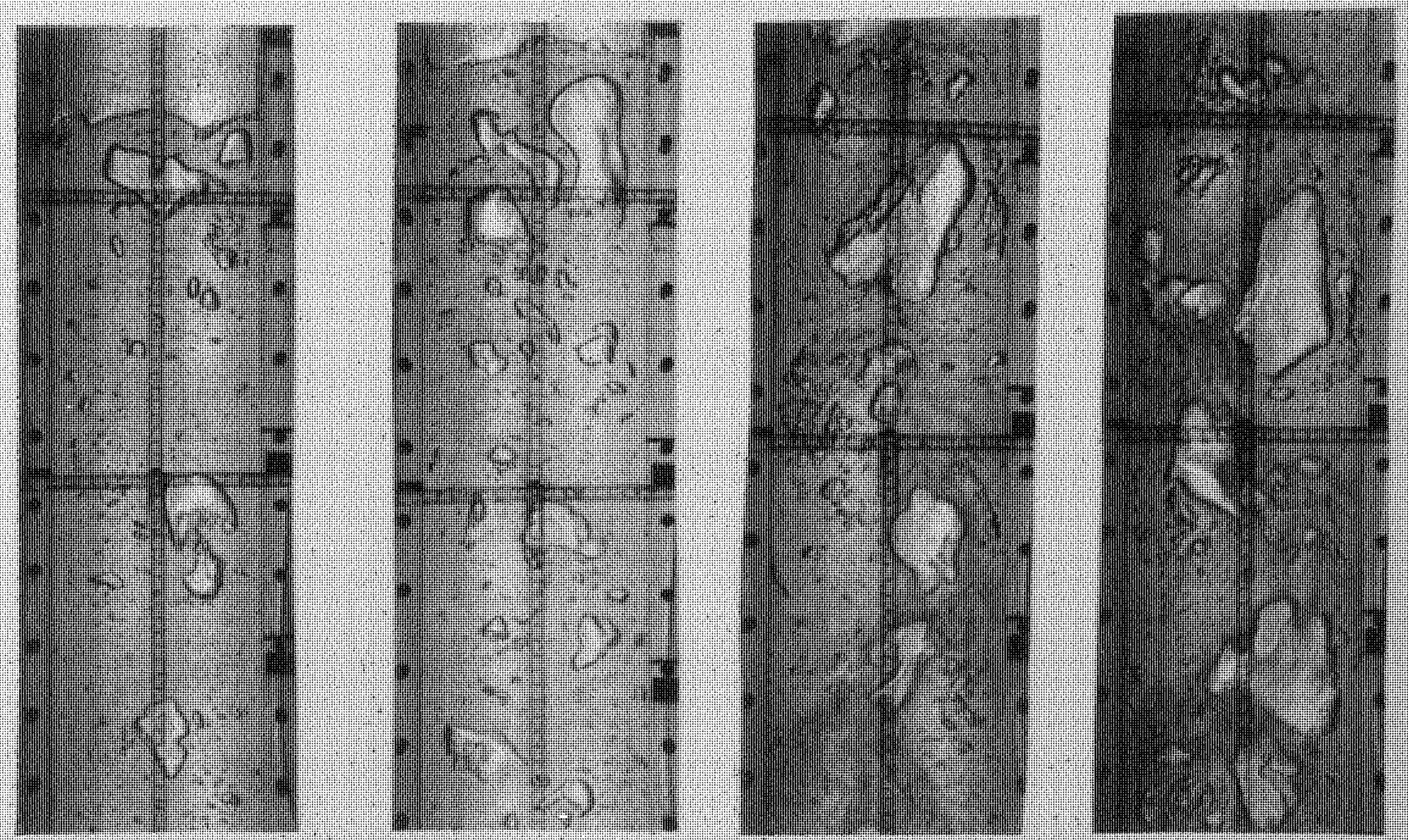
(ii) density ratio 2.21

	(a)	(b)	(c)	(d)
U_{sg} (cm/s)	3.10	11.45	15.23	19.21

Figure 8.8 contd.

(iii) density ratio 2.63

(a) (b) (c) (d)



U_{sg} (cm/s) 3.10 11.45 15.23 19.21

8.4 CIRCULATION PATTERNS IN THREE PHASE FLUIDISED SYSTEMS

Visualisation of the dynamic structure of flow was achieved by monitoring the liquid pathway within the system. In the present work, for clear photographic evidence to be obtained, dark coloured particles of ballotini 250μm (navy blue) were suspended in the system. Although the circulation patterns were clear to the naked eye when large particles were employed, they did not show well photographically. Observations were made easier in the case of low density particles if they were of large size.

The importance of liquid circulation has been previously recognised. Freedman and Davidson(1969), Reitema and Ottengraf (1970) and Hills(1975) have developed mathematical models to obtain liquid circulation velocities, which were later used by Lewis et al (1982) and Sharma et al (1976, 1982, 1983) in their heat and mass transfer models of bubble columns. A review of the relevant literature has been presented earlier in chapter 3.

The development of the circulation cells and subsequent "stable" circulation patterns was observed in the two-dimensional bubble column. The experimental arrangement was as illustrated earlier in figure 8.1.

The following variables were investigated:

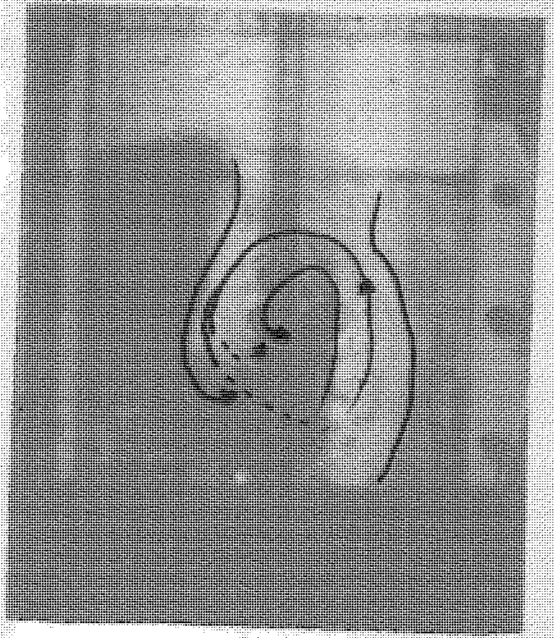
* total bed depth
* the superficial gas velocity
* solid properties

The circulation patterns were observed at three bed depths, H = 15, 30, 60 and 90cm above the distributor plate, corresponding to an aspect ratio (H/D) of 0.98, 1.93, 3.90, and 6.83 repectively. The results are presented as photographs, accompanied by sketches of the circulation pathways. The results were interpreted by comparison and integration with information reported in the literature, particularly the work of Freedman and Davidson (1969), Joshi and Sharma (1978, 1979, 1982), Chen et al (1989), and Davanathan et al (1990).

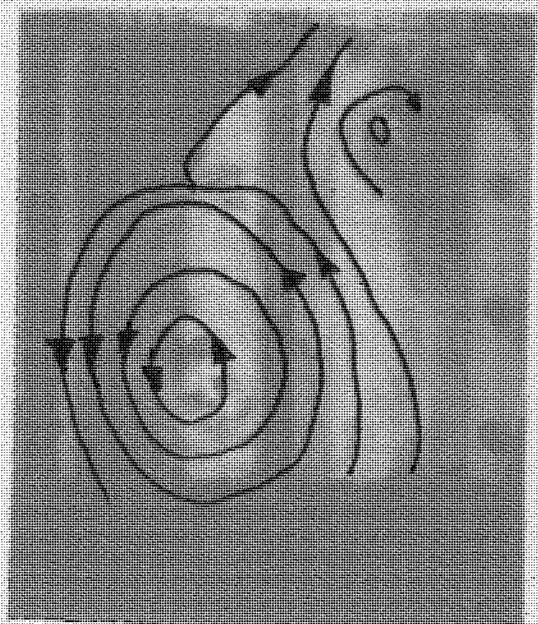
8.4.1 DEVELOPMENT OF CIRCULATION CELLS

The author regards the development of the circulation cells to be related to the effects observed when the aspect ratio is less than or equal unity. Figure 8.9 shows the photographic evidence obtained at this condition. The two-dimensional models of

Figure 8.9 : Single circulation cell at aspect ratio less or equal to one.

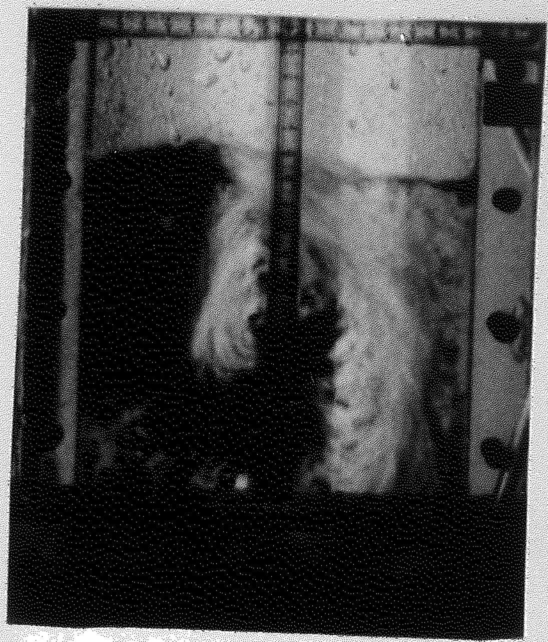


Usg = 11.45 cm/s



Usg = 19.21 cm/s

Figure 8.9 : Single circulation cell at aspect ratio less or equal to one.



Usg = 11.45 cm/s

Usg = 19.21 cm/s

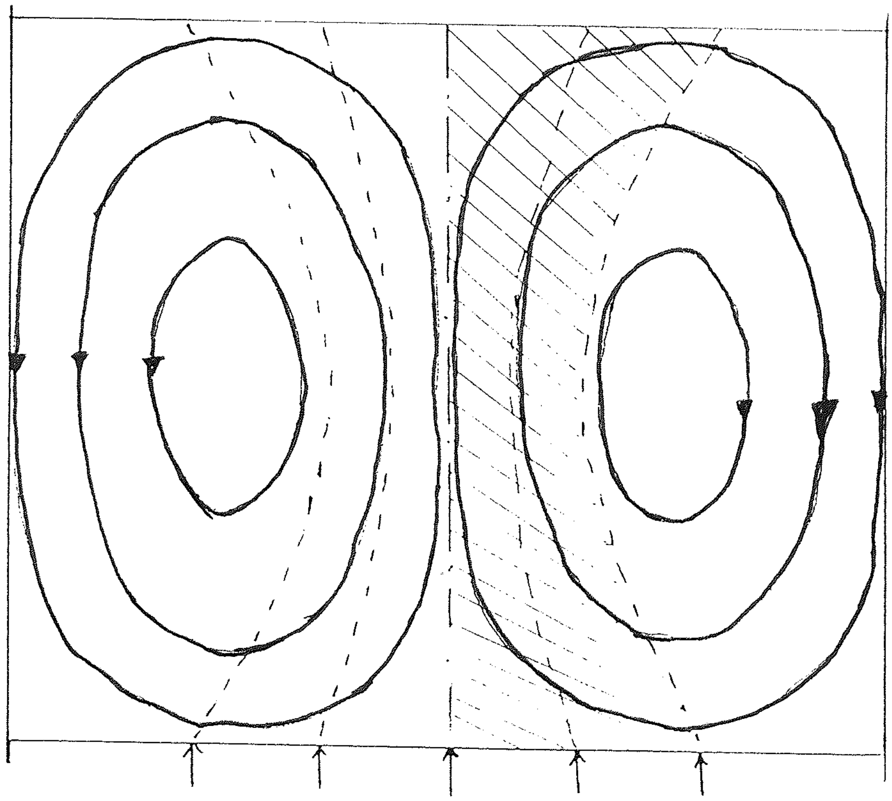


Figure 8.10 : Two-dimensional model of Freedman and Davidson showing the "gulf- stream" or "cooling tower" effect

Freedman and Davidson showing the "gulf-stream" or "cooling tower" effect with two vortex cells side by side, Figure 8.10, was observed. However, the pair of cells are short-lived, appearing and disappearing rapidly with a tendency to give a single circulation cell which alternated with the gulf-stream.

Chen et al (1989) provided photographic evidence for the existence of the two cells side by side at aspect ratios less or equal to one (H/D≤1). However, the material they used to track the liquid pathways was stationary, fixed to the sides of the column. In the author's work the suspended solids moved along the circulation pathways. Recently, Devanathan et al (1990), using a non-invasive computer automated radioactive particle tracking (CARPT) technique, observed a single recirculation cell with liquid ascending along the column center and descending along the wall. They concluded that this was the primary flow pattern, but found evidence of the existence of a small "secondary" cell at certain velocities. They observed two recirculating cells at gas velocities less than 5 cm/s.

8.4.2 MULTIPLE CIRCULATION CELLS

At higher aspect ratios, H/D>1, visual observations indicated a tendency towards a multiple cell pattern of vortices, in an alternate arrangement (or "staggered" arrangement - Chen et al 1989), in the axial direction. This arrangement was observed to be the stable hydrodynamic state within the bubble column at all operating conditions. The following typical characteristics were observed

* each circulation cell tended to position itself near the centre of the bubble column
* each cell exhibited similar circulation pathways (ie liquid flow was upward in the central direction and down ward near the column wall)
* the height of each cell in the axial direction was approximately the column width
* each cell occupied between two-thirds and the full column width when fully developed at high aspect ratios
* the number of the circulation cells increased with increasing bed level (or increasing aspect ratio).

Figures 8.11 and 8.12 show the typical characteristics of multiple circulation cells in a three-phase fluidised bubble column. The reader is also referred to figures 8.4 (ii) and 8.5.

Various multiple circulation cell models have been proposed by different workers, as summarised below.

Figure 8.11: Liquid circulation in a two-dimensional bubble column .
(high aspect ratio, H/D = 6.83)

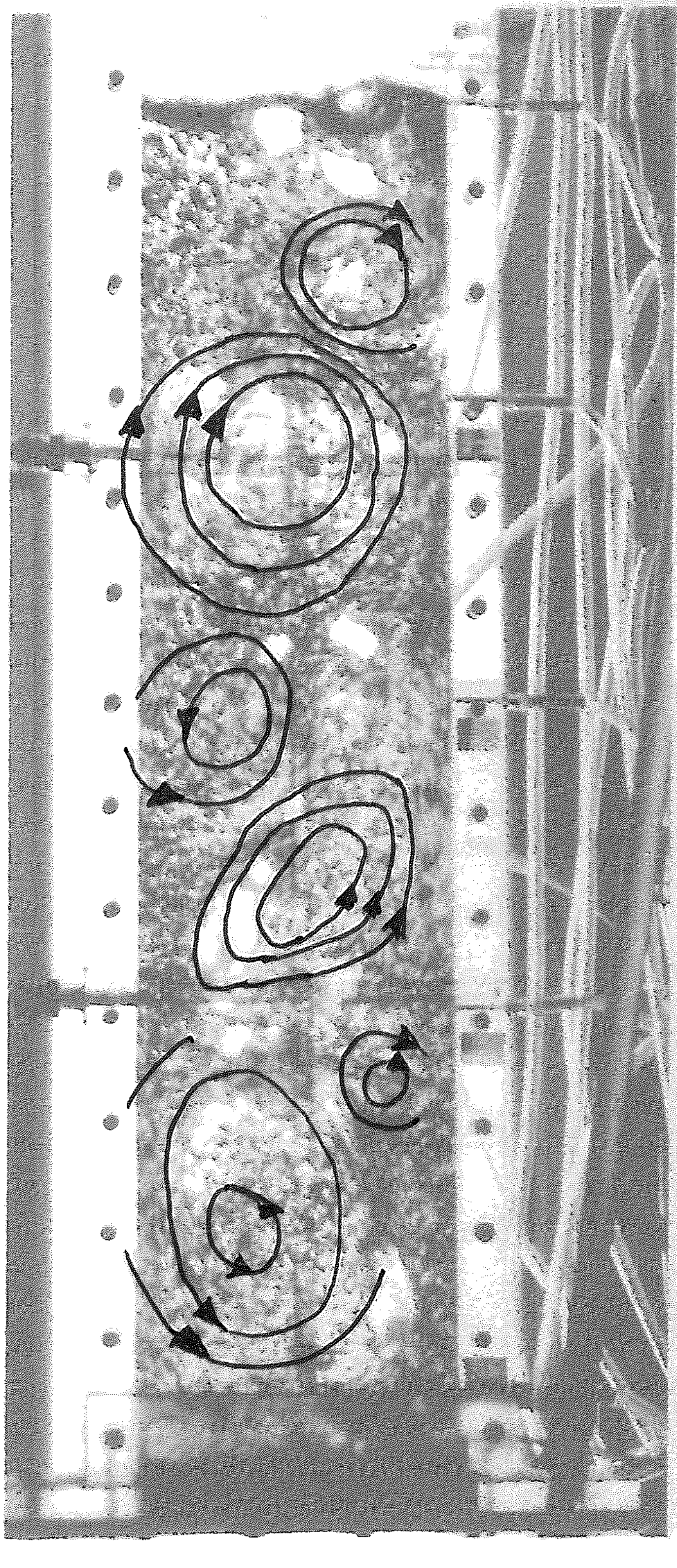


Figure 8.11: Liquid circulation in a two-dimensional bubble column .
(high aspect ratio, H/D = 6.83)

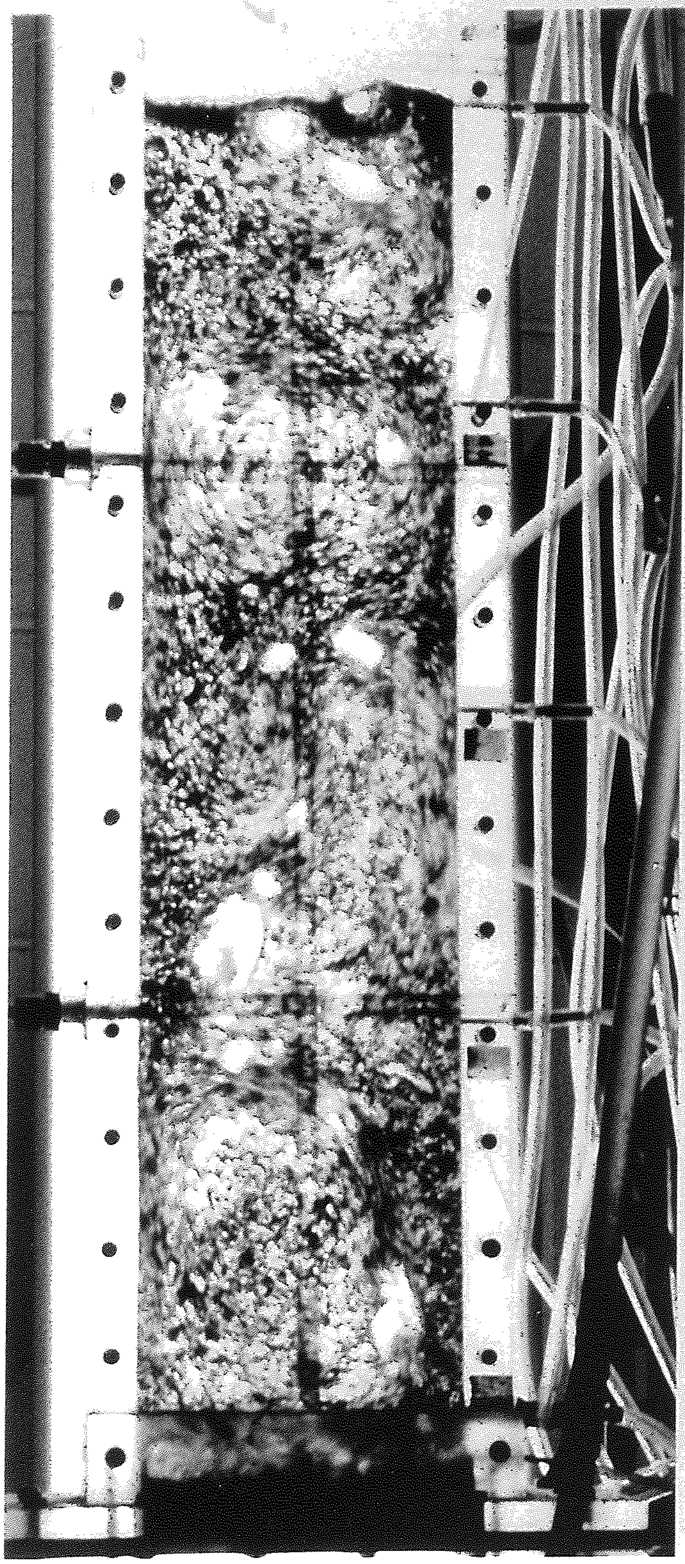
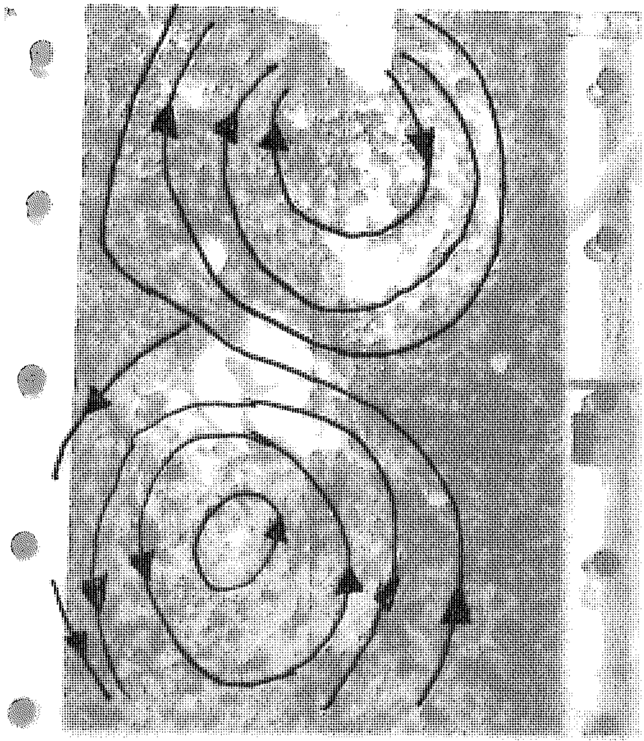


Figure [illegible] Staggered [illegible] multiple circulation ce[illegible]

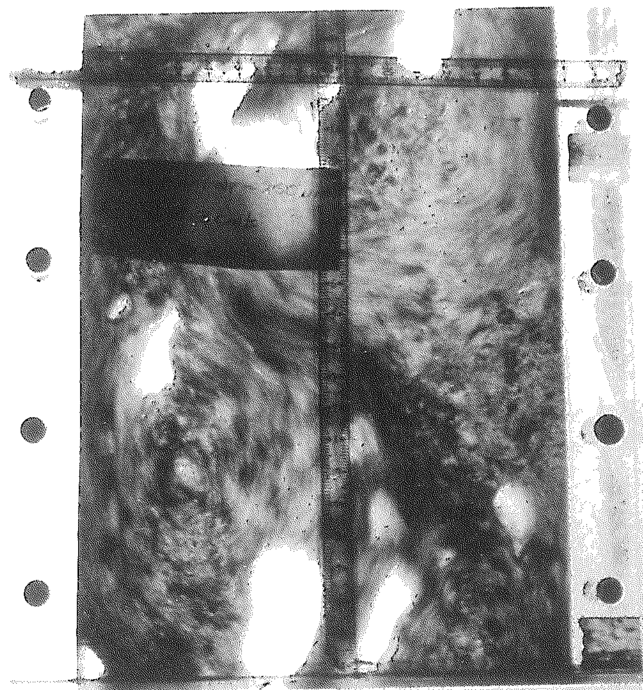


Usg = 11.45cm/s

Usg = 19.21cm/s

Figure 8.12 Staggered arrangement of multiple circulation cell

Usg = 11.45cm/s



Usg = 19.21cm/s

JOSHI AND SHARMA (1978, 1979, 1981)

They proposed that bubble columns contained multiple circulation cells in the axial direction arranged in the manner illustrated in figures 8.13a and b: this flow pattern has been criticised as being unrealistic by Van Der Akker and Rietema (1982). Zehner (1986) and Chen et al (1989) did not observe the circulation patterns of Joshi and Sharma: also, such patterns were not seen by the author in the present work.

CHEN ET AL (1989)

The observations made by Chen and his co-workers were similar to the results presented by the author. They observed a staggered arrangement of multiple circulation cells and related their findings to the work of Von Karman (1912) and the vortices shed behind cylinders (Karman vortex street). Von Karman showed that two infinite parallel rows of vortices cannot be stable if arranged as proposed by Joshi and Sharma, the only stable configuration being when the vortices are staggered, as observed in the present work.

8.4.3 EFFECT OF BED DEPTH

The effect of the bed depth was examined by varying the bed aspect ratio, (H/D). Generally, at high aspect ratios (H/D > 1), the circulation cells were stable, and there was no tendency to form a single vortex. The number of circulation cells appeared to increase with increasing aspect ratio, due to the occurence of secondary circulation cells, causing the cells to take more central position, occupying the width of the column. Figure 8.11 shows the liquid circulation in the bubble column, (high H/D ~ of 6.8).

8.4.4 EFFECT OF SUPERFICIAL GAS VELOCITY, Usg

Figure 8.12 shows the liquid circulation patterns observed at two gas velocities, Usg = 11.45 and 19.21 cm/s ~~when~~ with aspect ratio, (H/D >1). Earlier figure 8.9 gave a result for (H/D) <1. No variation was observed in the fully developed pattern when the aspect ratio was greater than unity. Each circulation cell characteristic remained the same. At high gas velocities and high aspect ratios the small "secondary" circulation cells occured occasionally, as indicated in figure 8.11.

The actual liquid circulation velocity increased, promoting the gulf stream when the aspect ratio was less than unity.

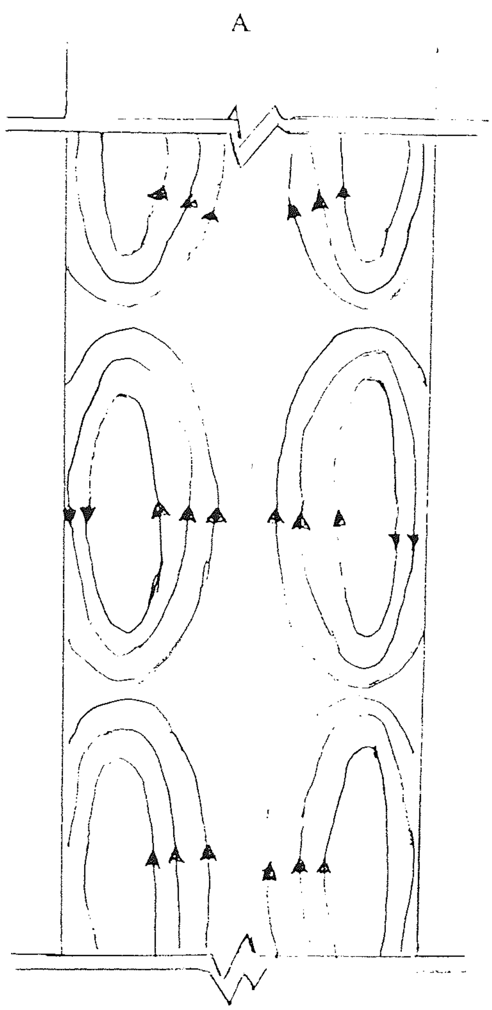


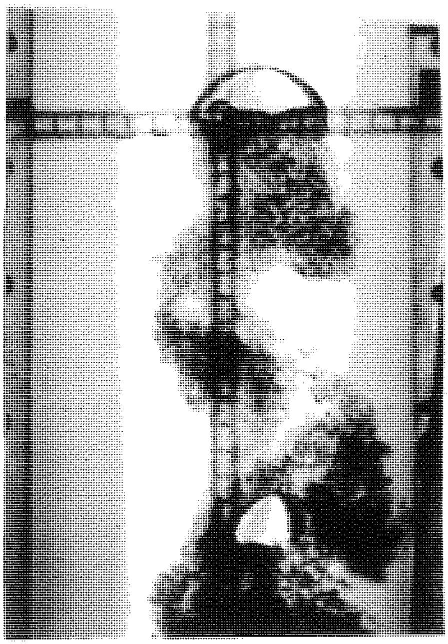
Figure 8.13: Multiple Circulation Cells Flow Pattern of Joshi and Sharma

A - interacting circulation cell model
B - non-interacting circulation cell model

Figure 8.14 : Solid particle flow pattern by bubble dynamics

air- water- ballotini (600μm; $\rho_p = 2.84$ g/cm^3)

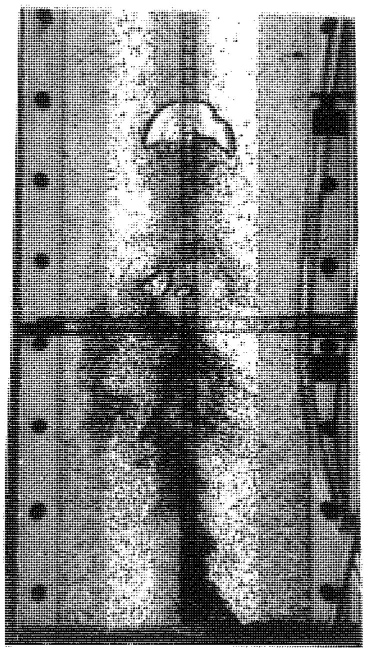
(a)



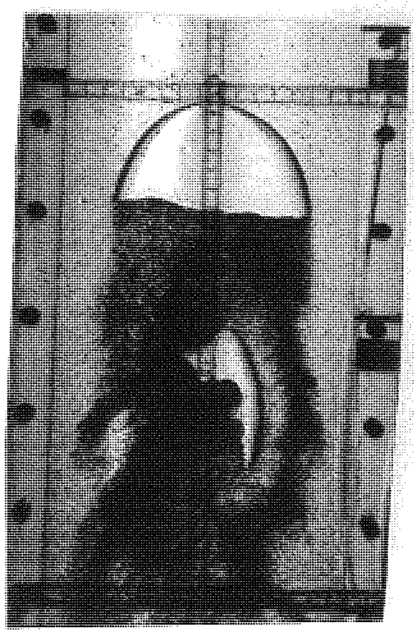
(b)

air-water-amberlite (600μm; $\rho_p = 1.20$ g/cm^3)

(c)



(d)



CHAPTER NINE

MIXING AND SEGREGATION IN THREE-PHASE FLUIDISED BUBBLE COLUMNS CONTAINING HIGH SOLID CONCENTRATIONS

9.1 INTRODUCTION

This chapter concerns the measurement of the degree of mixing in a three phase fluidised system consisting of two or more solid components differing in size or density or both, and examines the mechanism that gives rise to the steady state axial distributions of the solid components.

A solids mixing index was used to describe the degree of axial solid mixing in the bed. Furthermore, solid concentration and holdup profiles were used to predict the mixing states under the experimental operating conditions. Qualitatively, the relationships to flow regime and flow circulation pattern and associated physical mechanisms as observed and recorded in chapter 8 were used to provide further explanations.

Four binary particle mixtures were used (see Table 9.1). A general introduction and basic concepts relevant to this chapter have been discussed in chapter 2 and a review of the experimental studies can be found in chapter 3 (section 3.5). The equipment layout and operating procedures are described in chapter 4. Overlaps occur frequently within chapters 5, 6, 7 and 8, in the sections concerned with the discussion of experimental results. However efforts have been made to limit this chapter to the subject covered by the title.

The Mixing Index, (M)

The concept of the mixing index , M, first introduced by Rowe et al (1972) for liquid fluidised systems and later used by Fan et al (1987) (see chapter 2) has been re-defined to suit the author's requirement. Hence

$$\text{Mixing Index, } M = \bar{v} / v \qquad 9.1$$

where $\bar{v}$ is the volume of the "Measured Particle Component" in the sample taken over the top 25% of the total working volume, and v - is the volume of the "Measured Particle Component" in the entire system at the start of the experiment.

"Measured Particle Component"

To determine the particle component of the binary solid mixing ,the following guidelines were used:

1. If particle sizes are different but particle density is the same, the measured particle component is the larger size particle.
2. If particle sizes are similar but densities differ, then the measured particle is the heavier particle.

Table 9.1 Properties of binary solids mixtures used in this study

Binary Mixture	Component 1	Component 2	Size Ratio small / large	*Density Ratio heavy / light
m1	Ballotini	Ballotini	1: 2	1.03
m2	Styrocell	Styrocell	1: 2	1.11
m3	Amberlite	Diakon	1: 2	1.08
m4	Ballotini	Styrocell	1: 2	2.31

* Refer to Table 4.2 for the actual particle densities of the solids.

3. If both size and density differ, but the smaller particle is the heavier, then guideline 2 is followed; if the reverse is the case, guideline 1 is followed.

These guidelines were set on the basis of results obtained from preliminary experiments.

The Mixing Index, M, varies from 0 for the completely segregated state to 1 for the completely mixed state.

9.2 EXEPRIMENTAL PROCEDURE

Experiments were carried out both in the 2D- and 3D- bubble columns, following the operating procedures already described in chapter 4. The solids mixtures used are listed in Table 9.1. The measurement of the volume and concentration of individual particle components was carried out by the sampling method described in section 4.6.2. Caution was exercised to avoid preferential sampling of particle components at the sampling point nearest to the packed section, particularly with systems containing ballotini, and at the top of the column, where accumulation of small or lighter particles was possible, particularly at low gas velocities. Preferential sampling due to size difference between these two points was considered to be negligible.

9.3 EXPERIMENTAL RESULTS

9.3.1 EFFECT OF SUPERFICIAL GAS VELOCITY

Figure 9.1 shows typical results obtained for the effect of superficial gas velocity on the solids mixing index, M , in both the 2D and 3D bubble columns for solid binary mixtures m1 and m2, which had comparable size and density ratios.

For both binary mixtures, M increased with increasing superficial gas velocity to reach values close to 1. For the Ballotini mixture m1, the mixing index increased to give a typical S-shaped curve that was obtained for all binary mixtures containing Ballotini as one of the components.

The results obtained with binary mixture m2 were similar to those for binary mixture m3 (Amberlite / Diakon). For the binary mixture m4 (Ballotini / Styrocell), the results were similar to that obtained for binary mixture m1.

Figure 9.1: Effect of gas velocity on solid mixing index in 2D and 3D bubble columns

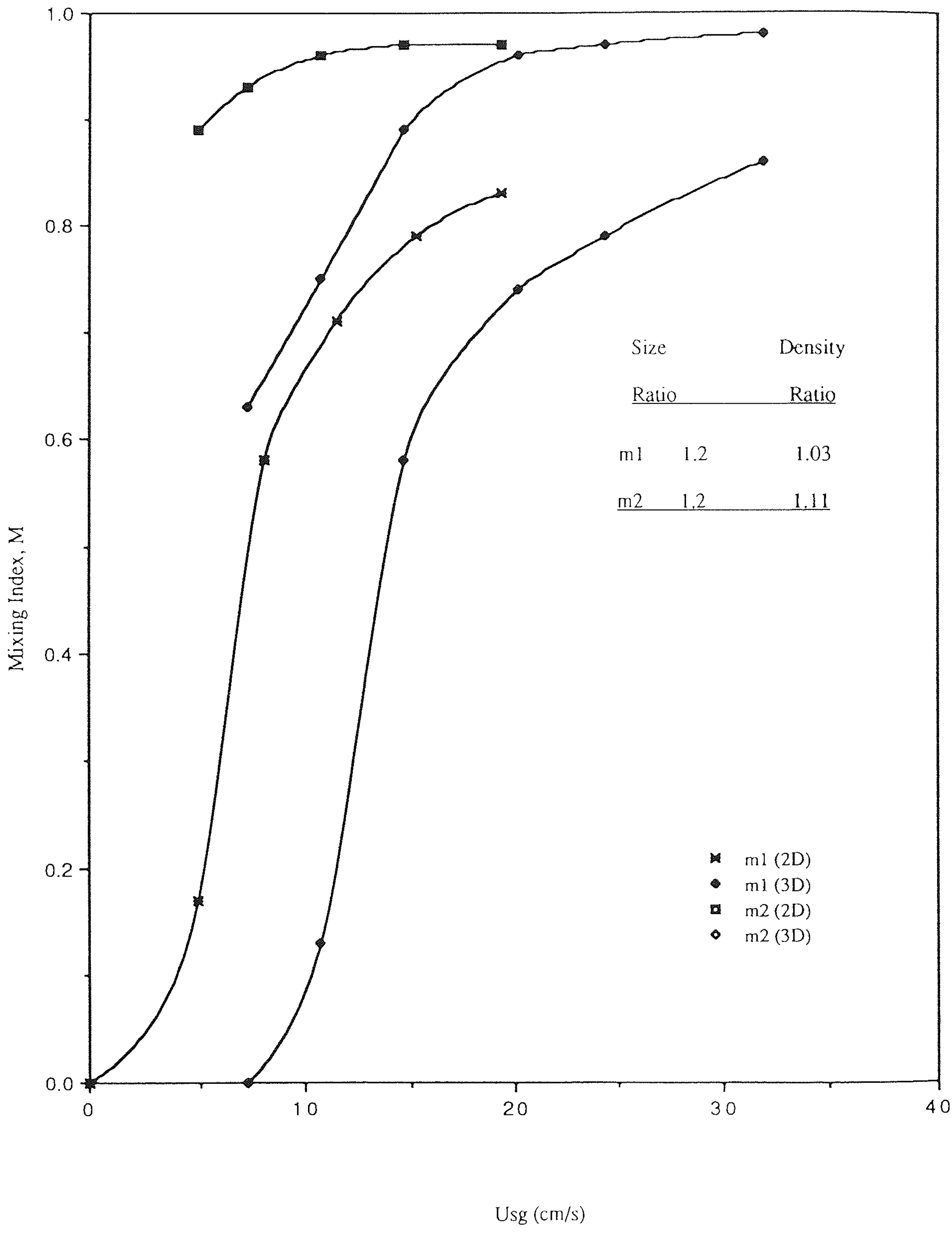
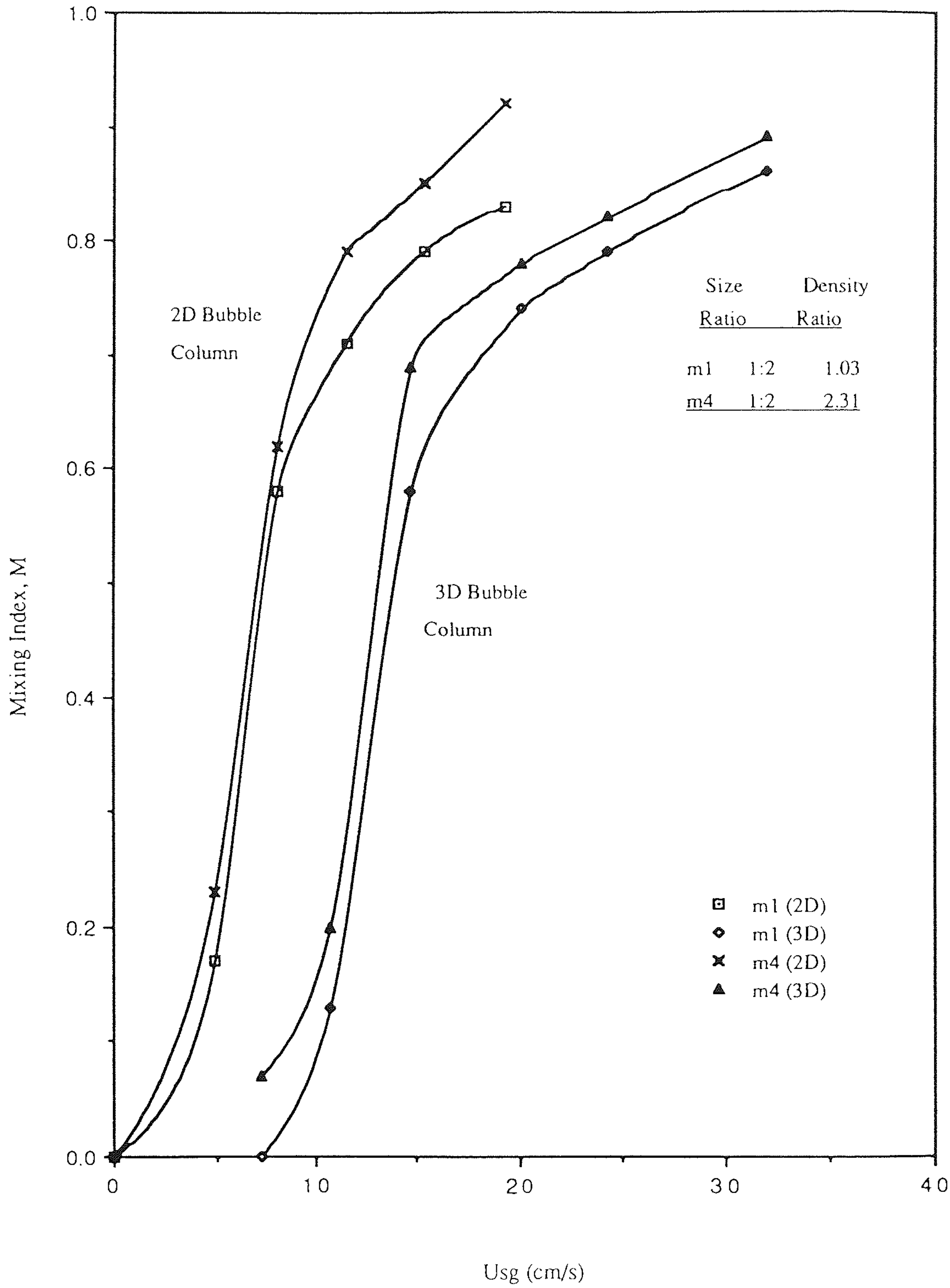


Figure 9.2: Effect of density ratio on mixing index



Therefore, with binary solid mixtures containing Ballotini as one of the components, a much higher gas velocity is needed to reach the same solid mixing index as that for other mixtures.

9.3.2 EFFECT OF DENSITY AND DENSITY RATIO

Figure 9.2 shows the results obtained when the effect of density ratio was explored for mixtures containing Ballotini as one of the components. The effect of density ratio appeared to be linked with the actual particle size of the measured particle component, namely Ballotini.

For binary mixture m1, the measured particle component was Ballotini of 1200μm. For binary mixture m4, the measured component was Ballotini of 600μm diameter. This could be the reason for the slightly higher mixing index obtained with mixture m4 for a given superficial gas velocity in both columns. This result is supported by the visual observations recorded in chapter 8. Photographs in Figures 8.3 illustrate the behaviour of Ballotini 600μm and 1200μm as single component solids in the three phase systems and those in Figure 8.6 reveal the behaviour of Ballotini as one of the components of binary mixtures with Styrocell at various size ratios. These two plates clearly indicate the degree of segregationwith a high fraction of the larger particles at the bottom of the column when particle density is comparable (Ballotini 600μm , Styrocell 1200μm).

9.3.3 EFFECT OF SIZE AND SIZE RATIO

The effect of particle size and size ratio on the mixing index is difficult to detach from that of density and density ratio. However, there is an indication that some sort of "trade-off" or interaction of these properties occurs.

Figures 9.3 and 9.4 illustrate some of the results obtained. In the case of binary particle mixtures m1 and m4, the measured particle components were Ballotini 1200μm and 600μm; Styrocell 1200μm was the other measured particle component (Figure 9.4 case 3).

The mixing index curve, in all cases, was moved slightly to the left with decreasing particle size at a given superficial gas velocity.

Figure 9.3: Effect of "measured particle component" size and size ratio

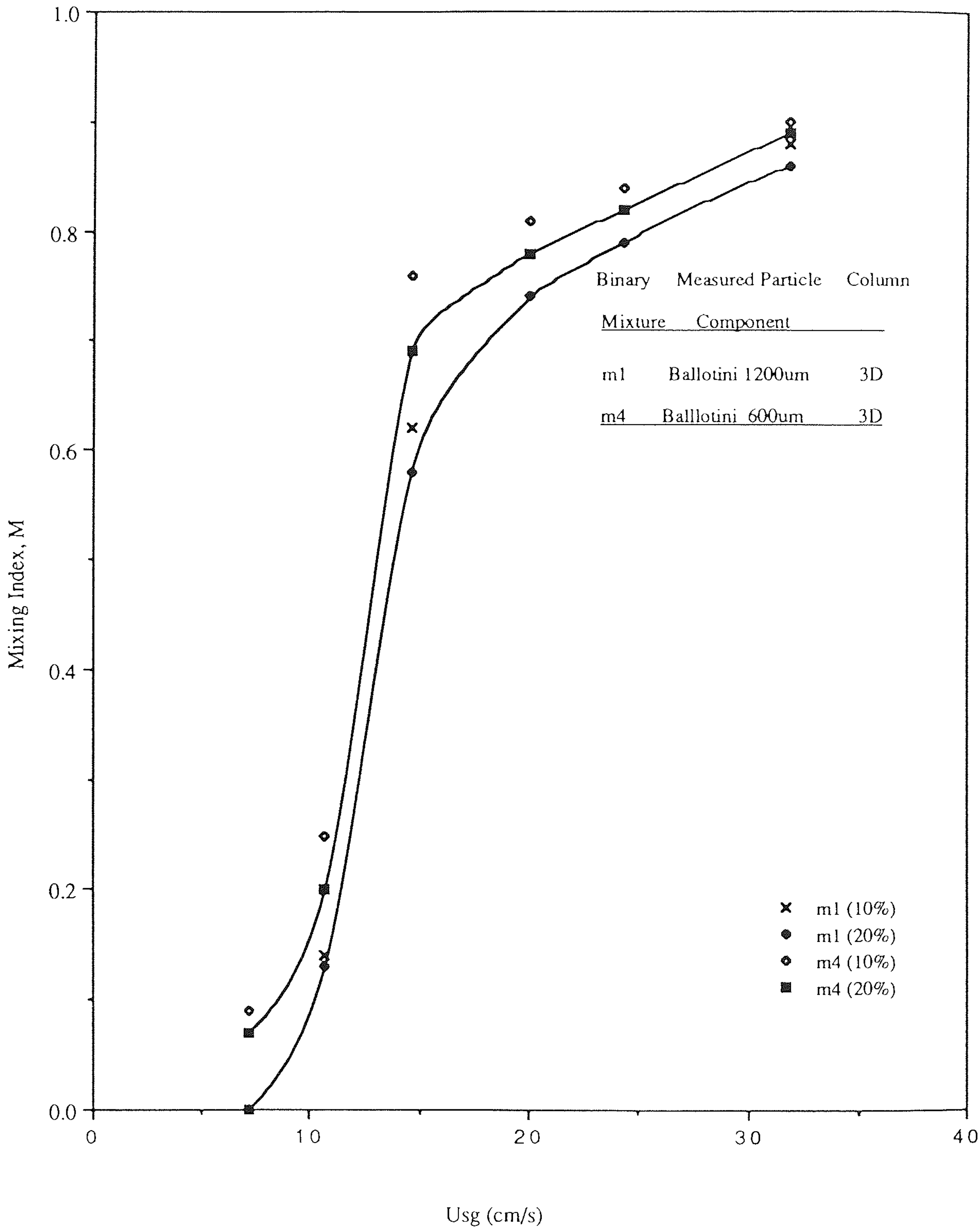
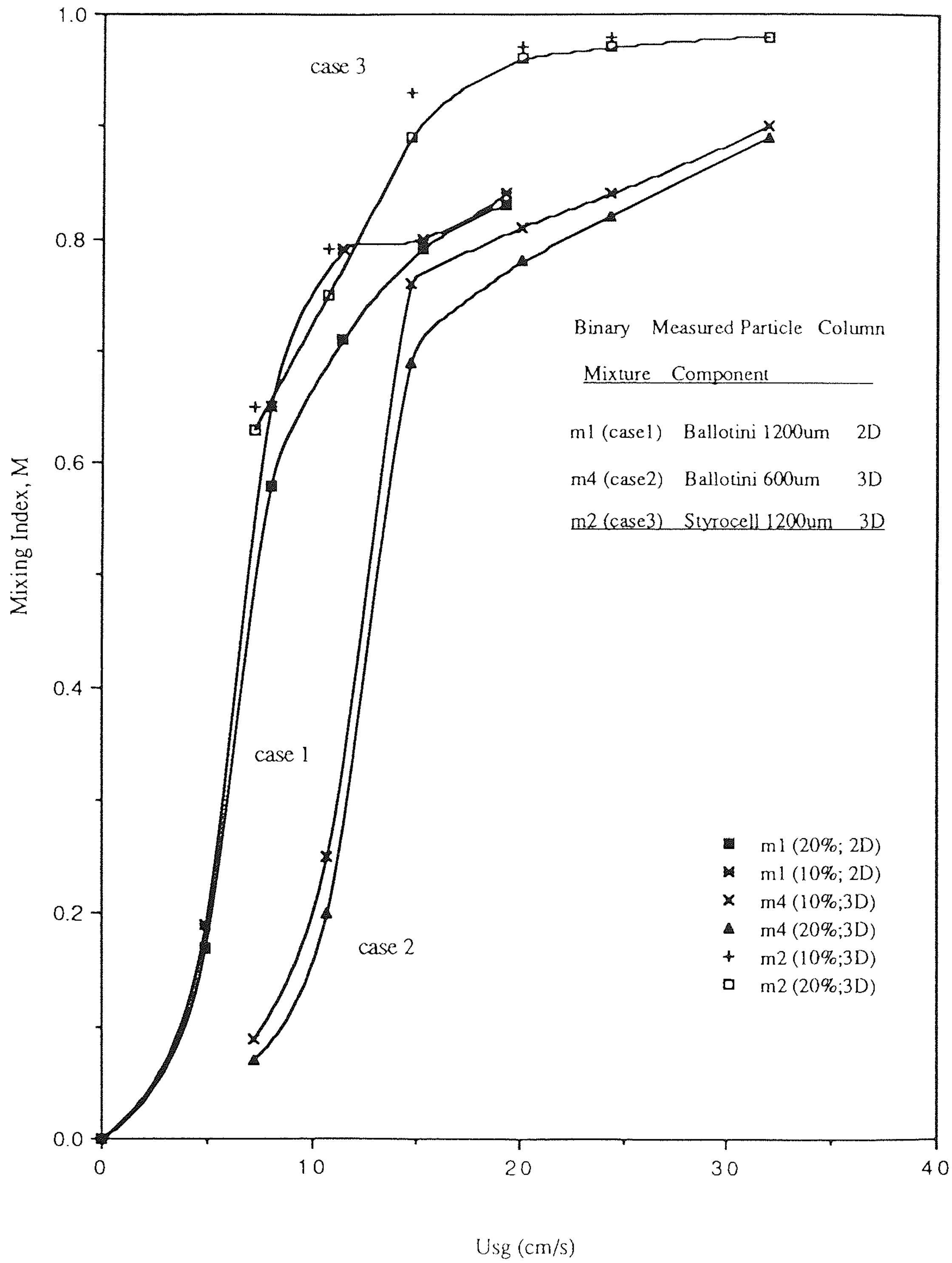


Figure 9.4: Effect of "trade-off" of properties on the mixing index



9.3.4 THE TRADE-OFF OF PROPERTIES

The direction in which the mixing index curve shifts was found to be dependent on the interaction of the physical properties of the measured particle component, initial concentration and column geometry. A reduction in concentration of 50% was found to increase the mixing index by up to 8% for a given superficial gas velocity; a greater mixing index was obtained in the 2D bubble column for a given measured particle component, (see Figure 9.4).

9.3.5 COLUMN GEOMETRY

From Figures 9.1 and 9.4, it can be concluded that a higher mixing index is obtained in the 2D bubble column (cf case 1 and case 2).

9.3.6 EFFECT OF GAS AND SOLID HOLDUPS

The variation in phase holdups reflects the solid concentration variation in the bed. Gas and solid holdups for systems containing binary solid mixtures are reported in chapter 6. Visual examination shows that the solid holdup distribution depends on the mixing state, which in turn depends on the flow conditions.

9.3.7 RESULTS OF QUALITATIVE STUDIES

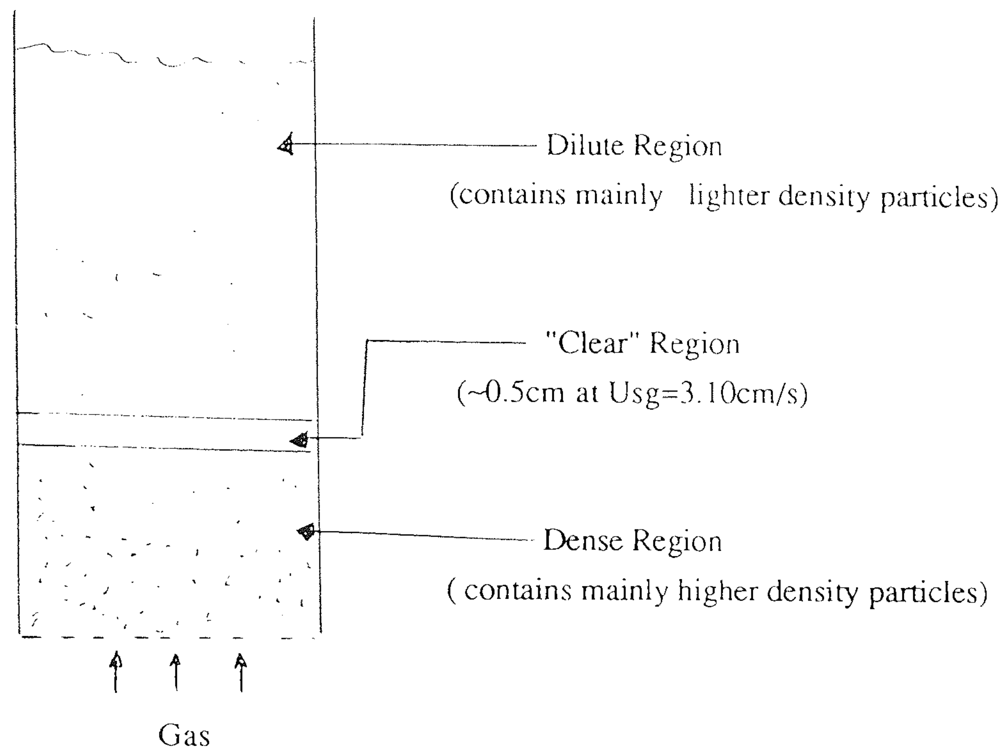
<u>Solids Distribution</u>

Except for systems containing Ballotini / Ballotini mixtures, the following observations were made. At low gas velocities (< 5 cm/s) a largely dilute region containing light particles (Styrocell, Amberlite, Diakon) was observed above a dense region containing Ballotini. There was a distinct strip or "clear" region, of up to 0.5cm, between the two regions. This region decreased on increasing the superficial gas velocity and was not visible beyond about 3.10 cm/s, giving rise to a uniformly distributed three phase fluidised bed containing proportions of each component of the binary particle mixtures with increasing gas velocity. Figure 9.5 provides schematic representation of this initial observation.

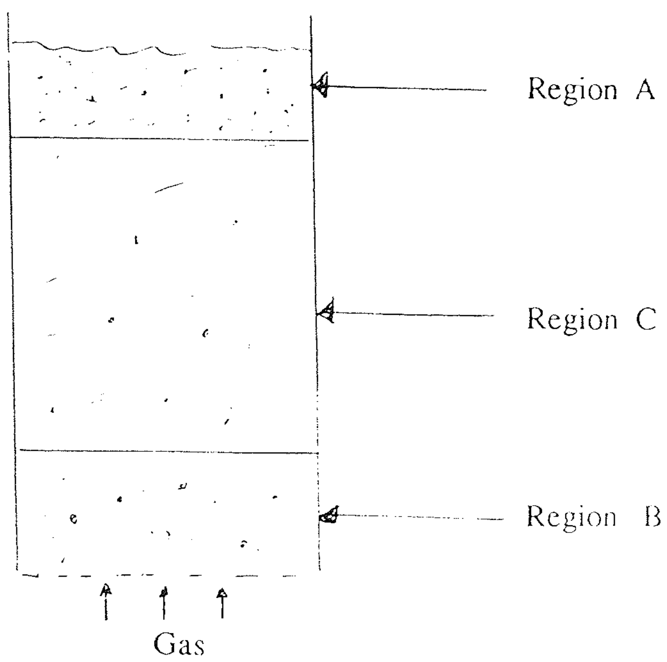
A close examination of the axial solids concentration distribution revealed three regions at high gas velocities (>5cm/s):

Figure 9.5: Schematic representation of bed regions in three phase fluidised bubble columns

1. Low Gas velocities (Usg < 5cm/s)



2. High Gas Velocites (Usg > 5cm/s)



Region A : at the top 20-25%of the bed. Here the concentrations of the large and small, or heavy and light, particles are constant : with higher concentrations of either the smaller or lighter components.

Region B: at the bottom 25% of the bed; This region was characterised by a constant concentration of each particle component , with a higher concentration of either larger or heavier particles.

Region C: In this region, located between region A and B , the concentration of each solid component showed variations axially.

Flow Regimes

Both the quantitative and qualitative observations correlate strongly with the flow regimes as recorded in chapter 8. Typically, at gas velocities above 5 cm/s, the bed was characterised by coalesced bubble flow, except for the case of mixtures containing Diakon as one of the components where significantly dispersed bubbly flow was observed. At higher gas velocities, when the system progresses to full churn-turbulent flow, marked solids mixing induced by the large bubbles was evident. Hence, the shape of the mixing curve and its characteristics can be explained in terms of the gas flow regimes and bubble properties.

Liquid Circulation

The circulation velocity of the liquid phase, induced by the relative velocity of the gas flow, plays an important role in solids mixing (or segregation) and solids distribution in the bed. A relatively low liquid velocity is observed at the bottom section of the bed corresponding to region B in Figure 9. 5. Surface erosion of the packed solids is activated by the liquid circulation cells and the shedding of the larger or heavier solids is also rapid leading to the establishment of region B. The velocity of the liquid circulation vortices reduces axially with the upward flow to the top of the column. The downward flow of solids, particularly near the column walls, is greater for heavier or larger particles, and gives rise to region A. The variation of the binary solids components in region C may be due, in part, to the observed interruption of liquid circulation cells by bubbles and slugs.

CHAPTER TEN: CONCLUSIONS, ACHIEVEMENTS, RECOMENDATION AND SUGGESTIONS FOR FUTHER WORK

10.1 CONCLUSIONS AND ACHIEVEMENTS

The objectives of the research study, as set out in section 1.2 have been achieved; the work is one of few studies to address the behaviour of true three-phase gas-liquid-solid fluidised bubble columns.

The following achievements and conclusions have been made.

10.1.1 PHASE HOLDUP

(1) gas and solid holdup have been studied and compared for systems containing single, binary and ternary solids particle mixtures

(2) similar trends in gas holdup exist for all systems containing single, binary and ternary particle mixtures

(3) generally, a reduction in gas holdup at any given superficial gas velocity will result due to the presence of solids when compared with a solid-free system (air-water)

(4) for all systems, gas holdup is decreased by increasing solid concentration and this is particularly significant for concentration increase from 0 to 15% v/v

(5) for air-water-single component systems (excluding Ballotini) at high superficial gas velocity, the dependency of gas holdup on concentration diminishes and values approach those for air-water

(6) the effect of concentration is only apparent if size and size ratio are similar for binary and ternary systems

(7) results with solid mixtures can be viewed in terms of the competition between physical properties studied, i.e size,density, concetration, and wettability, the "trade -off" of properties being a useful approach to explain this result

(8) very little or no effect on solid holdup is produced by particle size variation , except with systems containing Diakon in which an increase in particle size produced a noticeable but unexpected increase in average solid holdup

(9) solid holdup is sensitive to density and density ratio; particles of similar density have identical solid holdups

10.1.2 GAS FLOW REGIME

(1) In three-phase systems, at low superficial gas velocity, the bubbly flow regime is suppressed, due to the interacting influence of the solid present

(2) bubble coalescence and bubble breakup are strong functions of the gas velocity and axial position in the column; bubble coalescence occurs about the middle section and breakup takes place near the top and at the to interface in the case of the author's equipment

(3) increasing the solid concentration in three-phase operation delays the point at which the churn-turbulent state can be attained

(4) particle size does not have a significant effect on flow regime for solids of low density

(5) the effect of particles is significant for operations at low gas velocity where the bubbly regime is normally expected

(6) the effect of surface properties (solids "wettability") is not clearly established; however,some evidence of its existence is shown with systems containing Diakon and Amberlite as one of solid components

10.1.3 LIQUID AND SOLID CIRCULATION

(1) At low aspect ratios, $H/D \leq 1$, a dynamically stable circulation state consists of single circulation cell, alternating with the gulf-stream effect

(2) at high aspect ratios, $H/D>1$, multiple circulation cells are in "staggered" arrangement axially up the column

(3) each cell exhibits similar circulation pathways

(4) when fully developed , each cell occupies betwen two-thirds of, and the full column width

(5) solid circulation is dependent on the liquid circulation pattern,which in turn depends on the gas flow

10.1.4 MIXING AND SEGREGATION

(1) Guidelines for defining a mixing index involving the "measured particle component" have been established

(2) the mixing index increases with increasing superficial gas velocity to reach a value close to unity, when complete mixing can be assumed

(3) typically, S-shape curves are obtained for systems containing solid of high density (Ballotini); and levelling off to a constant value, are observed with systems of low density solids

(4) the direction in which the mixing index curve shifts is dependent on the interaction of the various physical properties

(5) the mixing index curve moves to the left with decreasing particle size, and a reduction of concentration by 50% from 20% volume by volume will produce up to 8% increase in the mixing index at a given gas velocity

(6) a degree of segregation occurs with systems containing a high fraction of large particles or heavier components at all gas velocities

(7) qualitatively, three mixing regions have been identified, based on solid concentration distributions (section 9.3.7 and figure 9.5)

10.2 RECOMMENDATIONS AND SUGGESTIONS FOR FURTHER WORK

The emphasis in the present work has been placed on physical and experimental concepts. It has been shown that the design of three-phase fluidised bubble columns is complicated by such factors as axial variations of holdups and distribution of solids of complex or mixed properties. There are very few data or equations for predicting the trends, and hence, the reactor performance under high fluid flow rates and solid loading, where these axial variation could be important. The present work has highlighted the following areas for further studies.

APPROACH

Experimental measurement of the aerated bed height by both visual and pressure gradient methods has its limitations, because the bed height fluctuates at high flow rate, and the measured pressure gradient is not particularly sensitive for systems with low density particles. However, other methods (see Shah (1982, Muroyama et al (1986, Olajuyigbe (1986)) could give rise to greater inaccuracy. The author recommends that by taking the average height in the positions within the band of fluctuations should give a more acceptable aerated bed height.

SOLID HOLDUP

The solid holdup was determined by using equation 5.2 ($\varepsilon_s = w_s / \rho\, A_{cs}\, H_f$), in which the element of axial variation has been ignored. Also, actual solid holdup decreases with increasing axial position. Therefore, it is recommended that the equation 5.2 should be modified to $\varepsilon_s\, dh = dw_s / \rho\, A_{cs}$.

Gas holdup is found to be more sensitive to solid holdup distribution in systems with high solid loading. Further work should result in correlations for the distribution of solid holdup throughout the entire bed, and its overall and local effects at the top and bottom 25% of the working volume. It is suspected that this could be crucial particularly in systems containing two or more solid components, where local conditions throughout the bed must be considered when designing such reactors.

FLOW PATTERNS

Observations of the flow patterns have raised doubts on the validity of most models available in the literature. Mathematical analysis of the "staggered" vortices to obtain more realistic models for prediction is necessary. Models that have been based on "gulf-stream " effects for columns with aspect ratio >1 could not be considered as accurate; this is also the case for aspect ratios <1, because of the unstable nature of flow, as observed and recorded in chapter 8.

Liquid circulation cells are a characteristic feature. All cells are identical and are relatively constant in their properties throughout the column. The experimental measurements of the height and width of fully developed cells are possible. Also, determination of solid holdup/solid concentration within each cell is possible by careful design of the sampling technique. It should be noted that the number of cells in the column is a strong function of the bed height, i.e the working volume. These could serve as important points in the development of predictive modelling equations.

MIXING AND SEGREGATION

In the absence of any conventional correlating approach for the systems used, the author believes that by taking mass balances, a statistical approach could be developed to provide a correlation for the mixing index which would cover the entire range of physical properties and operating conditions. Among the variables that should be considered are :

$$M = fn \{ U_{sg}, U_{sl}, \rho_{ratio}, V_{ratio}, h, \sigma, d_p, \}$$

Predictive equations could be useful in determining the transition stages between established mixing regions and possibly lead to a mixing state map for the systems used. With this in mind, further experimentation particularly in 3D bubble columns is suggested.

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APPENDIX A

Pressure correction for gas flowrate measurement

Superficial gas velocity, U_{sg}, is the volumetric flowrate at a given cross-section of the column divided by the cross-section area .

$$U_{sg} = Q/A \qquad \text{A1}$$

where Q is the gas flow rate, A = local sectional area

The gas flowrate was measured by taking readings from a rotameter incorporated into the airline. A rotameter chart was then used to estimate the flow rate, and as the mains pressure was different from atmospheric pressure a corresction was made.

Correction formula adopted is

$$Q_{\text{corrrected}} = Q_{\text{graph}} \sqrt{\frac{\text{Actual Area pressure(Main Pressure)}}{\text{Atmospheric pressure}}} \qquad \text{A2}$$

$Q_{corrected}$ is used in equation A1 in calculating the superfical gas velocity, U_{sg}

For 2-D bubble column

$$Q_{\text{Corrected}} = Q_{\text{graph}} \sqrt{\frac{10.0 + 14.7}{14.7}} \qquad \text{A3}$$

$$= Q_{\text{graph}} \times 1.3 \qquad \text{A4}$$

For 3-D bubble column

$$Q_{\text{corrected}} = Q_{\text{graph}} \sqrt{\frac{32 + 14.7}{14.7}} \qquad \text{A5}$$

$$= Q_{\text{graph}} \times 1.78 \qquad \text{A6}$$

Any variation in line pressure is accounted for accordingly.

For full derivation, refer to

Coulson, D.M and Richardson J.F "Chemical Enginering ". Vol 1, 3rd Ed, Pergamon. Press, Oxford (1977).

APPENDIX B

Table B1: Effect of Superficial Gas and Liquid Velocity on Gas Holdup in a 2-Dimensional Bubble Column

A. air-water-Amberlite (600μm; 10%v/v) in 2D column

	Gas Holdup, ε_g		
Usg(cm/s)	Usl=0cm/s	Usl=0.05cm/s	Usl=0.15cm/s
0.00	0.00	0.00	0.00
3.10	0.069	0.063	0.054
4.95	0.103	0.092	0.084
6.79	0.129	0.120	0.115
8.05	0.149	0.119	0.117
11.45	0.189	0.130	0.126
13.68	0.209	0.152	0.148
15.23	0.219	0.169	0.165
17.65	0.238	0.195	0.192
19.21	0.256	0.221	0.210

B air-water-Styrocell (600μm; 10%v/v) in 3D column

		Gas Holdup, ε_g		
Usg(cm/s)	Air-Water	Usl=0cm/s	Usl=0.05cm/s	Usl=0.15cm/s
0.00	0.00	0.00	0.00	0.00
5.10	0.138	0.070	0.060	0.053
7.21	0.187	0.089	0.079	0.069
10.69	0.214	0.121	0.109	0.095
14.62	0.224	0.153	0.138	0.121
20.01	0.253	0.179	0.159	0.142
24.30	0.285	0.201	0.181	0.169
28.78	0.311	0.228	0.203	0.178
31.82	0.336	0.239	0.212	0.201

Table B2: Effect of Solid Concentration on Gas holdup the 2-Dimensional Column

A. [Air-Water-Styrocell R751 (600μm)]

		Gas Holdup, ε_g	
Usg (cm/s)	Air-Water	10% v/v	20%v/v
0.00	0.00	0.00	0.00
3.10	0.093	0.063	0.067
4.95	0.135	0.095	0.107
6.79	0.164	0.126	0.139
8.05	0.172	0.151	0.158
11.45	0.200	0.178	0.195
13.68	0.217	0.202	0.219
15.23	0.227	0.218	0.230
17.65	0.244	0.250	0.255
19.21	0.264	0.250	0.255

B. [Air-Water-Styrocell R751 (1200μm)]

		Gas Holdup, ε_g	
Usg (cm/s)	Air-Water	10% v/v	20%v/v
0.00	0.00	0.00	0.00
3.10	0.093	0.054	0.063
4.95	0.135	0.088	0.098
6.79	0.164	0.112	0.121
8.05	0.172	0.127	0.136
11.45	0.200	0.166	0.175
13.68	0.217	0.188	0.201
15.23	0.227	0.202	0.212
17.65	0.244	0.214	0.228
19.21	0.264	0.233	0.241

C. [Air-Water-Diakon (600μm)]

Usg (cm/s)	Air-Water	Gas Holdup, ε_g 10% v/v	20%v/v
0.00	0.00	0.00	0.00
3.10	0.093	0.077	0.069
4.95	0.135	0.105	0.108
6.79	0.164	0.127	0.139
8.05	0.172	0.145	0.154
11.45	0.200	0.177	0.183
13.68	0.217	0.203	0.205
15.23	0.227	0.222	0.219
17.65	0.244	0.236	0.245
19.21	0.264	0.251	0.259

D. [Air-Water-Diakon (1200μm)]

Usg (cm/s)	Air-Water	Gas Holdup, ε_g 10% v/v	20%v/v
0.00	0.00	0.00	0.00
3.10	0.093	0.070	0.063
4.95	0.135	0.113	0.099
6.79	0.164	0.141	0.123
8.05	0.172	0.159	0.134
11.45	0.200	0.213	0.197
13.68	0.217	0.213	0.197
15.23	0.227	0.223	0.211
17.65	0.244	0.242	0.233
19.21	0.264	0.249	0.246

E. [Air-Water-Amberlite (600μm)]

Usg (cm/s)	Air-Water	Gas Holdup, ε_g 10% v/v	20%v/v
0.00	0.00	0.00	0.00
3.10	0.093	0.069	0.077
4.95	0.135	0.103	0.114
6.79	0.164	0.129	0.139
8.05	0.172	0.149	0.155
11.45	0.200	0.189	0.193
13.68	0.217	0.209	0.216
15.23	0.227	0.219	0.225
17.65	0.244	0.238	0.241
19.21	0.264	0.256	0.260

F. [Air-Water-Amberlite (1200μm)]

Usg (cm/s)	Air-Water	Gas Holdup, ε_g 10% v/v	20%v/v
0.00	0.00	0.00	0.00
3.10	0.093	0.067	0.065
4.95	0.135	0.102	0.101
6.79	0.164	0.126	0.12
8.05	0.172	0.144	0.143
11.45	0.200	0.182	0.175
13.68	0.217	0.204	0.198
15.23	0.227	0.215	0.212
17.65	0.244	0.238	0.228
19.21	0.264	0.248	0.242

Table B3: Effect of Solid Concentration on Gas holdup in the 3-Dimensional Column

[Air-Water-Ballotini (1200μm)]

	Gas Holdup, ε_g		
Usg (cm/s)	Air-Water	10% v/v	20%v/v
0.00	0.00	0.00	0.00
5.10	0.138	0.070	0.070
7.21	0.187	0.089	0.089
10.69	0.214	0.121	0.119
14.62	0.224	0.153	0.146
20.01	0.253	0.179	0.172
24.30	0.285	0.201	0.195
28.78	0.311	0.228	0.220
31.82	0.336	0.239	0.235

Table B4: Effect of Solid Size on Gas Holdup in a 2-D Bubble Column

A. Air-Water-Ballotini (10%v/v)

		Gas Holdup, ε_g	
Usg (cm/s)	Air-Water	600μm	1200μm
0.00	0.000	0.000	0.000
3.10	0.093	0.067	0.065
4.95	0.135	0.102	0.101
6.79	0.164	0.126	0.124
8.05	0.172	0.144	0.143
11.45	0.200	0.182	0.175
13.68	0.217	0.204	0.198
15.23	0.227	0.215	0.212
17.65	0.244	0.238	0.228
19.21	0.264	0.248	0.242

B. Air-Water-Ballotini (20%v/v)

		Gas Holdup, ε_g	
Usg (cm/s)	Air-Water	600μm	1200μm
0.00	0.000	0.000	0.000
3.10	0.093	0.055	0.056
4.95	0.135	0.090	0.089
6.79	0.164	0.114	0.110
8.05	0.172	0.126	0.122
11.45	0.200	0.161	0.155
13.68	0.217	0.183	0.179
15.23	0.227	0.197	0.189
17.65	0.244	0.215	0.205
19.21	0.264	0.225	0.212

C. Air-Water-Styrocell(10%v/v)

Usg (cm/s)	Air-Water	Gas Holdup, ε_g 600μm	1200μm
0.00	0.000	0.000	0.000
3.10	0.093	0.063	0.054
4.95	0.135	0.095	0.088
6.79	0.164	0.126	0.112
8.05	0.172	0.151	0.127
11.45	0.200	0.178	0.166
13.68	0.217	0.202	0.188
15.23	0.227	0.218	0.202
17.65	0.244	0.237	0.214
19.21	0.264	0.250	0.233

D. Air-Water-Styrocell(20%v/v)

Usg (cm/s)	Air-Water	Gas Holdup, ε_g 600μm	1200μm
0.00	0.00	0.00	0.00
3.10	0.093	0.067	0.063
4.95	0.135	0.107	0.098
6.79	0.164	0.139	0.121
8.05	0.172	0.158	0.136
11.45	0.200	0.195	0.175
13.68	0.217	0.219	0.201
15.23	0.227	0.230	0.212
17.65	0.244	0.243	0.228
19.21	0.264	0.255	0.241

Table B5: Effect of Solid Density on Gas Holdup

A. Air-Water-Ballotini (10%v/v)
Air-Water-Styrocell (10%v/v) 2D Bubble Column; 1200μm

Usg (cm/s)	Air-Water	Gas Holdup, ε_g Ballotini	Styrocell
0.00	0.00	0.00	0.00
3.10	0.093	0.063	0.054
4.95	0.135	0.091	0.088
6.79	0.164	0.117	0.112
8.05	0.172	0.133	0.127
11.45	0.200	0.170	0.166
13.68	0.217	0.185	0.188
15.23	0.227	0.201	0.202
17.65	0.244	0.221	0.214
19.21	0.264	0.225	0.233

B. Air-Water-Ballotini (20%v/v)
Air-Water-Styrocell (20%v/v) 2D Bubble Column; 1200μm

Usg (cm/s)	Air-Water	Gas Holdup, ε_g Ballotini	Styrocell
0.00	0.00	0.00	0.00
3.10	0.093	0.056	0.063
4.95	0.135	0.089	0.098
6.79	0.164	0.110	0.121
8.05	0.172	0.122	0.136
11.45	0.200	0.155	0.175
13.68	0.217	0.179	0.201
15.23	0.227	0.189	0.212
17.65	0.244	0.205	0.228
19.21	0.264	0.212	0.241

C. Air-Water-Styrocell (10%v/v)
Air-Water-Diakon (10%v/v) 2D Bubble Column; 1200μm

Usg (cm/s)	Gas Holdup, ε_g Air-Water	Styrocell	Diakon
0.00	0.00	0.00	0.00
3.10	0.093	0.054	0.070
4.95	0.135	0.088	0.113
6.79	0.164	0.112	0.141
8.05	0.172	0.127	0.159
11.45	0.200	0.166	0.196
13.68	0.217	0.188	0.213
15.23	0.227	0.202	0.222
17.65	0.244	0.214	0.242
19.21	0.264	0.233	0.249

D. Air-Water-Styrocell (20%v/v)
Air-Water-Diakon (20%v/v) 2D Bubble Column; 600μm

Usg (cm/s)	Gas Holdup, ε_g Air-Water	Styrocell	Diakon
0.00	0.00	0.00	0.00
3.10	0.093	0.063	0.063
4.95	0.135	0.098	0.099
6.79	0.164	0.121	0.123
8.05	0.172	0.136	0.134
11.45	0.200	0.175	0.176
13.68	0.217	0.201	0.197
15.23	0.227	0.212	0.211
17.65	0.244	0.228	0.233
19.21	0.264	0.241	0.246

E. Air-Water-Amberlite (10%v/v)
Air-Water-Diakon (10%v/v)

Usg (cm/s)	Gas Holdup, ε_g Air-Water	Amberlite	Diakon
0.00	0.00	0.00	0.00
3.10	0.093	0.069	0.077
4.95	0.135	0.103	0.105
6.79	0.164	0.129	0.127
8.05	0.172	0.149	0.145
11.45	0.200	0.189	0.177
13.68	0.217	0.209	0.205
15.23	0.227	0.219	0.222
17.65	0.244	0.238	0.236
19.21	0.264	0.256	0.251

F. Air-Water-Amberlite (20%v/v)
Air-Water-Diakon (20%v/v)

Usg (cm/s)	Gas Holdup, ε_g Air-Water	Amberlite	Diakon
0.00	0.00	0.00	0.00
3.10	0.093	0.077	0.069
4.95	0.135	0.114	0.108
6.79	0.164	0.139	0.139
8.05	0.172	0.155	0.154
11.45	0.200	0.193	0.183
13.68	0.217	0.216	0.205
15.23	0.227	0.225	0.219
17.65	0.244	0.241	0.245
19.21	0.264	0.260	0.253

Table B6: Effect of superficial gas and liquid velocity on solid holdup

Usg (cm/s)	Uls=0.25cm/s	Solid Holdup, Usl=0.75cm/s (HT=50cm)	ε_g Usl=0.75cm/s (HT=75cm)
3.10	0.053	0.043	0.042
4.95	0.048	0.036	0.034
6.79	0.041	0.031	0.027
8.05	0.038	0.029	0.023
11.45	0.036	0.023	0.018
13.68	0.033	0.019	0.015
15.23	0.031	0.018	0.012
17.65	0.030	0.016	0.009
19.21	0.029	0.014	0.008

2D Bubble Column; Ballotini 1200μm (10%v/v)

Table B7: Effect of solid concentration on solid holdup

Usg (cm/s)	Solid Holdup			
	Amberlite(10%)	Diakon (10%)	Amberlite (20%)	Diakon(20%)
3.10	0.054	0.054	0.106	0.107
4.95	0.052	0.052	0.101	0.103
6.79	0.050	0.051	0.098	0.099
8.05	0.049	0.050	0.097	0.096
11.45	0.047	0.048	0.092	0.094
13.68	0.046	0.046	0.090	0.092
15.23	0.045	0.045	0.089	0.090
17.65	0.044	0.044	0.087	0.087
19.21	0.043	0.043	0.085	0.086

2D Bubble Column; 600μm both cases

<u>B</u>

Usg (cm/s)	Solid Holdup			
	Ballotini(10%)	Styrocell(10%)	Ballotini(20%)	Styrocell (20%)
3.10	0.060	0.053	0.119	0.105
4.95	0.057	0.051	0.115	0.101
6.79	0.056	0.050	0.112	0.097
8.05	0.055	0.048	0.110	0.095
11.45	0.052	0.047	0.106	0.091
13.68	0.051	0.045	0.103	0.088
15.23	0.050	0.044	0.101	0.087
17.65	0.049	0.042	0.099	0.086
19.21	0.048	0.042	0.098	0.084

Table B8: Effect of solid particle size on solid holdup

A air-water-Ballotini (10%v/v) 2D Bubble Column;

Usg (cm/s)	Solid Holdup	
	Ballotini 600μm	Ballotini 1200μm
0.00	0.100	0.100
3.10	0.060	0.061
4.95	0.057	0.059
6.79	0.056	0.057
8.05	0.055	0.056
11.45	0.052	0.054
13.68	0.051	0.053
15.23	0.050	0.052
17.65	0.049	0.051
19.21	0.048	0.050

B
air-water-Diakon (10%v/v) 2D Bubble Column;

Usg (cm/s)	Solid Holdup	
	Diakon 600μm	Diakon 1200μm
0.00	0.100	0.100
3.10	0.054	0.062
4.95	0.052	0.059
6.79	0.051	0.057
8.05	0.050	0.056
11.45	0.048	0.053
13.68	0.046	0.052
15.23	0.045	0.052
17.65	0.044	0.050
19.21	0.043	0.050

APPENDIX C

Table C1: Effect of concentration and on gas holdup in a system containing binary particle mixtures

2D Bubble Column

	Gas Holdup				
Usg(cm/s)	M6a (10%)	M6a(20%)	M1a(10%)	M1a(20%)	Air-Water
0.00	0.00	0.00	0.00	0.00	0.00
3.10	0.062	0.072	0.055	0.051	0.093
4.95	0.096	0.098	0.082	0.088	0.135
6.79	0.122	0.122	0.109	0.112	0.164
8.05	0.139	0.138	0.122	0.126	0.172
11.45	0.176	0.171	0.150	0.161	0.200
13.68	0.196	0.190	0.172	0.178	0.217
15.23	0.199	0.204	0.180	0.187	0.227
17.65	0.219	0.223	0.197	0.202	0.244
19.21	0.241	0.239	0.211	0.213	0.264

Table C2: Effect of concentration ratio on gas holdup in a system containing binary particle mixture (2D Bubble Column)

Styrocell /Diakon m ixtures (10%v/v)
[Similar size and mixture ratio and reversed case for both]

Usg (cm/s)	Gas Holdup Binary mixture M4b (1: 2)	Binary Mixture M4c (2:1)
0.00	0.00	0.00
3.10	0.064	0.061
4.95	0.102	0.103
6.79	0.116	0.137
8.05	0.138	0.161
11.45	0.169	0.196
13.68	0.190	0.222
15.23	0.203	0.233
17.65	0.213	0.245
19.21	0.231	0.257

Table C3: Effect of size ratio on gas holdup in a system containing binary particle mixture (2D bubble column)

Binary Mixtures M1a, M1b and M1c
[Mixture Ratio 1:1] (10%v/v)

		Gas Holdup		
Usg (cm/s)	Size Ratio:	1:2	2:1	1: 1
0.00		0.00	0.00	0.00
3.10		0.058	0.062	0.050
4.95		0.084	0.090	0.082
6.79		0.111	0.113	0.109
8.05		0.124	0.134	0.122
11.45		0.154	0.165	0.150
13.68		0.174	0.189	0.172
15.23		0.197	0.199	0.180
17.65		0.211	0.219	0.197
19.21		0.226	0.229	0.211

Table C4: Effect of density and density ratio on gas holdup in a system containing binary particle mixtures

A. Size and Particle Mixture Ratio 1: 2 (10%v/v)

2D Bubble Column;

Usg (cm/s)	Gas Holdup	
Density Ratio:	M2b (2.6)	M5b (1.02)
0.00	0.00	0.00
3.10	0.064	0.057
4.95	0.092	0.086
6.79	0.127	0.111
8.05	0.142	0.129
11.45	0.175	0.161
13.68	0.199	0.183
15.23	0.208	0.196
17.65	0.222	0.210
19.21	0.237	0.223

B. Size and Particle Mixture Ratio 1: 1

Usg (cm/s)	Gas Holdup	
Density Ratio:	M1a (2.4)	M6a (1.07)
0.00	0.00	0.00
3.10	0.055	0.062
4.95	0.082	0.096
6.79	0.109	0.122
8.05	0.122	0.139
11.45	0.150	0.176
13.68	0.172	0.196
15.23	0.180	0.199
17.65	0.197	0.219
19.21	0.211	0.241

Table C5: Effect of total concentration on solid holdup in a system containing binary particle mixture

2D Bubble Column

Usg(cm/s)	Solid Holdup			
	M6a (10%)	M6a(20%)	M1a(10%)	M1a(20%)
3.10	0.056	0.112	0.048	0.096
4.95	0.055	0.110	0.047	0.094
6.79	0.053	0.106	0.046	0.092
8.05	0.052	0.104	0.045	0.090
11.45	0.051	0.102	0.043	0.086
13.68	0.049	0.098	0.042	0.084
15.23	0.049	0.098	0.041	0.082
17.65	0.048	0.096	0.040	0.080
19.21	0.047	0.094	0.039	0.078

Table C6: Effect of concentration ratio on solid holdup in systems containing binary particle mixture

A. Binary Mixture M2b; Size Ratio 1: 2 (10%v/v)

2D Bubble Column

Usg(cm/s)	Solid Holdup		
	Binary Mixture, M2b	Ballotini	Diakon
3.10	0.062	0.020	0.042
4.95	0.060	0.020	0.040
6.79	0.058	0.019	0.039
8.05	0.057	0.019	0.038
11.45	0.055	0.018	0.037
13.68	0.053	0.018	0.035
15.23	0.053	0.017	0.036
17.65	0.052	0.017	0.035
19.21	0.051	0.017	0.034

B. Binary Mixture M2c; Size Ratio 2: 1

2D Bubble Column

Usg(cm/s)	Solid Holdup		
	Binary Mixture, M2c	Ballotini	Diakon
3.10	0.059	0.042	0.017
4.95	0.058	0.041	0.017
6.79	0.056	0.039	0.017
8.05	0.054	0.038	0.016
11.45	0.052	0.037	0.015
13.68	0.051	0.036	0.015
15.23	0.049	0.035	0.014
17.65	0.049	0.035	0.014
19.21	0.048	0.034	0.014

Table C7: Effect of size and size ratio on solid holdup in systems containing binary particle mixture

A. Binary Mixture M5b Ratio 2:1; Size Ratio 1: 2 (10%v/v)

2D Bubble Column

	Solid Holdup		
Usg(cm/s)	Binary Mixture, M5b	Styrocell	Amberlite
3.10	0.052	0.035	0.017
4.95	0.051	0.034	0.017
6.79	0.049	0.033	0.016
8.05	0.048	0.033	0.015
11.45	0.047	0.032	0.015
13.68	0.045	0.031	0.014
15.23	0.044	0.030	0.014
17.65	0.044	0.029	0.015
19.21	0.042	0.029	0.013

B. Binary Mixture M5c Ratio 2:1; Size Ratio 2:1

2D Bubble Column

	Solid Holdup		
Usg(cm/s)	Binary Mixture, M5c	Ballotini	Diakon
3.10	0.051	0.033	0.018
4.95	0.049	0.032	0.017
6.79	0.048	0.031	0.017
8.05	0.046	0.030	0.016
11.45	0.045	0.029	0.016
13.68	0.044	0.029	0.015
15.23	0.043	0.028	0.015
17.65	0.042	0.027	0.015
19.21	0.041	0.027	0.014

APPENDIX D

Table D1: Effect of concentration on gas holdup in a system containing ternary particle mixtures

2D Bubble Column

Size and mixture ratio 1:1:1

Usg(cm/s)	Gas Holdup MT1(10%)	MT1(20%)	Air-Water
3.10	0.053	0.052	0.093
4.95	0.079	0.081	0.135
6.79	0.105	0.108	0.164
8.05	0.119	0.123	0.172
11.45	0.148	0.156	0.200
13.68	0.170	0.175	0.217
15.23	0.178	0.185	0.227
17.65	0.195	0.200	0.244
19.21	0.210	0.212	0.264

Table D2: Effect of concentration ratio on gas holdup in a system containing ternary particle mixture (2D Bubble Column)

Similar size ratio ; Total concentration: 10%v/v

Usg (cm/s)	Gas Holdup Mixture MT1 1: 1:1	Mixture MT2 1:1:2	Mixture MT3 2:2:1
3.10	0.052	0.062	0.060
4.95	0.081	0.104	0.089
6.79	0.108	0.136	0.114
8.05	0.123	0.160	0.132
11.45	0.156	0.197	0.164
13.68	0.175	0.223	0.187
15.23	0.185	0.235	0.202
17.65	0.200	0.244	0.218
19.21	0.212	0.258	0.234

Table D3: Effect of size ratio on gas holdup in a system containing ternary particle mixture (2D bubble column)

[Mixture Ratio 1:1:1]

Total concentration 20%

		Gas Holdup	
Usg (cm/s)	Size Ratio:	MT1 1:2: 1	MT3 2:1:1
3.10		0.059	0.062
4.95		0.086	0.090
6.79		0.113	0.113
8.05		0.127	0.134
11.45		0.158	0.165
13.68		0.177	0.189
15.23		0.201	0.199
17.65		0.216	0.219
19.21		0.230	0.229

Table D4: Effect of density and density ratio on gas holdup in a system containing ternary particle mixtures

Size and Particle Mixture Ratio 1: 1:1

Total concentration 10%v/v

2D Bubble Column

Usg (cm/s)		Gas Holdup	
	Density Ratio:	MT1 2.56: 1: 1	MT3 2.58 : 1 : 1
0.00		0.00	0.00
3.10		0.053	0.060
4.95		0.079	0.093
6.79		0.105	0.120
8.05		0.119	0.137
11.45		0.148	0.174
13.68		0.170	0.193
15.23		0.178	0.197
17.65		0.195	0.214
19.21		0.211	0.239

Table D5: Effect of total concentration and concentration ratio on solid holdup in a system containing ternary particle mixture

2D Bubble Column

Usg(cm/s)	Solid Holdup					
	MT3 10%	MT3 20%	MT2 10%	MT2 20%	MT1 10%	MT1 20%
3.10	0.054	0.097	0.056	0.112	0.048	0.096
4.95	0.047	0.095	0.055	0.110	0.047	0.094
6.79	0.046	0.093	0.053	0.106	0.046	0.092
8.05	0.045	0.092	0.052	0.104	0.045	0.090
11.45	0.043	0.090	0.051	0.102	0.043	0.086
13.68	0.042	0.086	0.049	0.098	0.042	0.084
15.23	0.041	0.084	0.049	0.098	0.041	0.082
17.65	0.040	0.082	0.048	0.096	0.040	0.080
19.21	0.039	0.080	0.047	0.094	0.039	0.078

Table D6: Effect of size and size ratio on solid holdup in systems containing ternary particle mixture

2D Bubble Column ; (10%v/v)

Usg(cm/s)	Solid Holdup	
	ternary mixture, MT1 2:2:1	ternary mixture, MT2 1:1:2
3.10	0.052	0.058
4.95	0.051	0.055
6.79	0.049	0.052
8.05	0.048	0.050
11.45	0.047	0.048
13.68	0.045	0.046
15.23	0.044	0.043
17.65	0.044	0.042
19.21	0.042	0.040

Table D7: Effect of density and density ratio on solid holdup in systems containing ternary particle mixture

2D Bubble Column; (10%v/v)

Usg(cm/s)	Density Ratio:	Solid Holdup MT1 2.56: 1: 1	MT3 2.58 : 1 : 1
3.10		0.052	0.054
4.95		0.051	0.053
6.79		0.049	0.052
8.05		0.048	0.050
11.45		0.047	0.048
13.68		0.046	0.046
15.23		0.045	0.044
17.65		0.044	0.043
19.21		0.042	0.041

APPENDIX E

Table E1: Effect of gas velocity on solid mixing index in 2D and 3D bubble column

Usg (cm/s)	Mixing index			
	m1(10%) 2D	m1(20%) 3D	m2(10%) 2D	m2(20%) 3D
0.00	0.00		0.09	
4.95	0.14		0.25	
7.21		0.00		0.07
8.05	0.62		0.76	
10.69		0.13		0.20
11.45	0.78		0.81	
14.62		0.58		0.69
15.23	0.82		0.84	
19.21	0.88		0.90	
20.01		0.74		0.78
24.30		0.79		0.82
31.82		0.86		0.89

Table E2: Effect of density ratio on mixing index

Usg (cm/s)	Mixing index			
	m1(2D)	m1(3D)	m4(2D)	m4(3D)
0.00	0.00		0.00	
4.95	0.17		0.23	
7.21		0.00		0.07
8.05	0.58		0.62	
10.69		0.13		0.20
11.45	0.71		0.79	
14.62		0.58		0.69
15.23	0.79		0.85	
19.21	0.83		0.92	
20.01		0.74		0.78
24.30		0.79		0.82
31.82		0.86		0.89

Table E3: Effect of "Measured Particle Component" size and size ratio

3D Bubble Column

Usg (cm/s)	Mixing index			
	m1(10%)	m1(20%)	m4(10%)	m4(20%)
0.00				
4.95				
7.21	0.00	0.00	0.09	0.07
8.05				
10.69	0.14	0.13	0.25	0.20
11.45				
14.62	0.62	0.58	0.76	0.69
15.23				
19.21				
20.01	0.78	0.74	0.81	0.78
24.30	0.82	0.79	0.84	0.82
31.82	0.88	0.86	0.90	0.89

Table E4: Effect of "Trade-off" of properties on the mixing index

Usg (cm/s)	Mixing index					
	m1 (20%;2D)	m1 (10%;2D)	m4 (10%;3D)	m4 (20%;3D%)	m2 (10%;3D)	m2 (20%;3D)
0.00	0.00	0.00				
4.95	0.17	0.19				
7.21			0.09	0.07	0.65	0.63
8.05	0.58	0.65				
10.69			0.25	0.20	0.79	0.75
11.45	0.71	0.79				
14.62			0.76	0.69	0.93	0.89
15.23	0.79	0.80				
19.21	0.83	0.84				
20.01			0.81	0.78	0.97	0.96
24.30			0.84	0.82	0.98	0.97
31.82			0.90	0.89	0.98	0.98