Design Parameters Estimations for Bubble Column Reactors

Gas-liquid as well as gas-liquid-solid bubble column reactors are extensively used in the chemical industry. This review evaluates the present state of the art for the estimation of "nonadjustable" parameters in bubble column reactors. All the pertinent literature concerning these parameters is discussed and the discussion is followed by relevant recommendations for their predictions. Literature on the modified bubble columns has been incorporated. Finally, the inadequacies of the data in some areas of practical importance have been pointed out and the recommendations for future work are outlined.

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SCOPE

It is well known that bubble column reactors have a wide range of applications such as absorption, catalytic slurry reactions, bioreactions, coal liquefaction etc. These reactors are preferred because of simplicity of operation, low operating costs, and ease with which liquid residence time can be varied. The vast literature makes it impossible to cover all the aspects of bubble column reactors in one review; therefore, we limit ourselves in this review to the parametric estimations in bubble column reactors. The only unified review on bubble columns was published by Mashelkar (1970). Since then, many articles have been published on different aspects of bubble columns, except for the estimations of non-adjustable parameters. This paper attempts to bring about a more complete and up-to-date review of this subject matter.

In the introduction, a comprehensive list of processes in which bubble columns are used is presented. Bubble dynamics and flow regimes indirectly influence the scaleup and design of bubble column reactors; and hence, these aspects are discussed initially along with relevant recommendations. Nonadjustable parameters like phase holdup, gas-liquid interfacial area, interfacial mass transfer coefficients, dispersion coefficients and heat transfer coefficients have a direct bearing on the problems associated with the design of the bubble column reactors. A critical analysis of the available literature on each of these parameters is presented in conjunction with the recommendations for their estimation. Wherever possible, the literature on modified bubble columns pertaining to these parameters, is also outlined. Finally, suggestions for the future work on bubble columns are offered.

CONCLUSIONS AND SIGNIFICANCE

Over the past two decades, an extensive effort has been directed to the scaleup of bubble column reactors. Though easy to use, bubble columns are difficult to design because of the complexity of flow characteristics, and their unknown behavior under different sets of design parameters such as diameter, height, etc. The present review indicates that there are numerous methods available to evaluate the non-adjustable parameters; few of which are based on extensive collection of data.

For water-like less viscous fluids, flow regime characteristics can be depicted with help of Richardson and Zaki's (1954) correlation. Unfortunately, no systematic effort has been made to study the effect of fluid properties or the nature of the gas distributor on the flow regime characteristics.

For estimating bubble diameters in bubble columns and jet reactors, the general equation of Calderbank (1967) (Eq. 3.1) is recommended. Also, Akita and Yoshida's (1974) correlation (Eq. 3.2) is applicable in the churn turbulent regime for less effective spargers (single and multiorifice spargers and perforated plates), and aqueous systems. Although, this correlation predicts the dependency of d_{vs} on the column diameter; for column diameter greater than 0.3 m, d_{vs} becomes independent of column diameter. For liquids of low viscosity and for bubbly flow, the guideline developed on the basis of Berghman's (1973) diagram for the dynamic equilibrium diameter should be taken into consideration. The bubble rise velocity can be estimated by the method of Clift et al. (1978) (Eq. 3.6).

The gas holdup is often very sensitive to the kind of liquid media, and the number of correlations proposed indicates that no single unified equation is available. One can therefore conclude that, the easily measurable physico-chemical properties such as density, viscosity and interfacial tension are not sufficient to describe the holdup data in bubble columns. The correlations of Akita and Yoshida (1973) and Hikita et al. (1980) are based on relatively large amounts of data for aeration with single and multinozzle spargers, and their application is recommended if the systems under consideration are similar. These equations are also applicable for three-phase systems if the slurry behaves as a pseudohomogenous fluid. Due to the lack of sufficient gas holdup data in organic media and liquid mixtures, actual measurements in 0.1–0.15 m diameter column are recommended.

Akita and Yoshida's (1974) (Eq. 5.3) correlation can be used to estimate a conservative value of interfacial area a, especially for less effective (but widely used) spargers. In general, the equation by Nagel et al. (1979) (Eq. 5.8) can be recommended. It has been proved to be valid for bubble columns aerated by

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two-phase nozzle spargers and other kinds of gas-liquid contactors. For non-Newtonian highly viscous media, Eq. 5.9 of Schumpe and Deckwer (1981) can be applied. For estimation of accurate values of interfacial area a, an absorption-reaction system which gives low conversion, i.e., in which bubble column can be operated as differential reactor, should be used. Sulfite oxidation is particularly recommended as an appropriate method.

For determination of k_La, the correlation of Akita and Yoshida (1973), which applies to columns with less effective spargers, would give a conservative estimate. In general, equations of type 6-a.2 (Deckwer et al., 1974; Kastanek, 1977) can be recommended with n = 0.8, but the constant b is very sensitive to the kind of sparger and the liquid medium. The correlation of Deckwer et al. (1981d) can be recommended for slug flow in non-Newtonian media. The effect of presence of solids on k_La depends on various parameters and no correlation is available at present time. If the particles are completely fluidized, k_La is little affected, otherwise k_La decreases in the presence of solids. For organic liquid medium and electrolyte solutions, there is a necessity of conducting some experiments with the specified system in 0.1 to 0.15 m diameter columns. k_La determination in tall bubble columns should be based on measurements of concentration profiles which should preferably be analyzed by using the axial dispersion model.

The correlations of Calderbank and Moo-Young (1961), and Hughmark (1967) which give similar predictions can be recommended for the estimation of k_L. No correlation is available for k_G and hence no recommendation can be given. The correlation of Sänger and Deckwer (1981) which covers a wide range of Schmidt and Reynolds number is recommended for determination of k_S.

Axial liquid-phase dispersion coefficient can be estimated with the help of correlations of Deckwer et al. (1974) (Eq. 7.1) or Joshi (1980) (Eq. 7.3); both of which describe the measured data in water like low viscous media equally well. Reliable liquid phase dispersion coefficient values for highly viscous and non-Newtonian media are not available in the literaure. For the estimation of gas phase dispersion coefficients, the correlation of Mangartz and Pilhofer (1980) (Eq. 7.12) is recommended. Solid dispersion coefficients can be calculated by the equation proposed by Kato et al. (1972a) (Eq. 7.14).

For calculation of the heat transfer coefficients in bubble columns, a correlation of Deckwer (1980) (Eq. 8.1) is recommended which applies to gas-liquid dispersion and to gas-liquid-solid systems if the particles are smaller than 120 μm. Data for suspensions with larger particles and higher particle concentrations (> 20% wt.) are not available in the literature.

The most important "adjustable" parameter which affects all of the above "nonadjustable" parameters is the column diameter. It can be concluded from the available literature that the effect of diameter on all these parameters (except the dispersion coefficient) becomes insignificant for $D_C \geq 0.15$ m, for less viscous liquids. For highly viscous ($\mu_L > 0.02$ Pa·s) liquids, this effect should become insignificant for $D_C \geq 0.3$ m. These values of column diameter are independent of other liquid properties.

Of all the parameters discussed in the following sections, it is believed that mass transfer coefficient is the most important design variable followed by gas holdup and axial dispersion coefficient. Heat transfer characteristics become important only if the reaction is highly exothermic or endothermic.

Finally, there are many regions in bubble column reactors where the hydrodynamic, mixing, and transport characteristics are not clearly understood. This paper outlines some of these areas and makes some recommendations for future study on this subject.

INTRODUCTION

Bubble columns are contactors in which a discontinuous gas phase in the form of bubbles moves relative to a continuous phase. The continuous phase can be a liquid or a homogeneous slurry. Bubble columns can be single staged or multistaged, batch or continuous, and can be operated cocurrently or countercurrently. Various types of bubble column reactors and their modifications are shown in Figure 1.1. Although some reviews concerning different aspects of bubble columns have appeared in the literature (Shah et al., 1978; Joshi and Shah, 1981; Shah and Deckwer, 1981); a unified review concerning the parametric estimations has not been published in the last decade.

The main advantages in using bubble columns compared to other multiphase contactors are:

1. Less maintenance is necessary due to the absence of moving parts.
2. Higher values of effective interfacial areas and overall mass transfer coefficients can be obtained.
3. Higher heat transfer rates per unit volume of the reactors can be achieved.
4. Solids can be handled without any errosion or plugging problems.
5. Less floor space is occupied and bubble column reactors are less costly.
6. Slow reactions can be carried out due to high liquid residence time.

Considerable backmixing in both phases, high pressure drop and bubble coalescence can be disadvantageous in some cases. Bubble columns have been modified in many ways to suit particular applications; e.g., loop reactors are used in the ICI pressure cycle fermenter and its deep shaft modification (Hines, 1978). For certain reactions like air oxidations of organic and inorganic compounds, where high gas component conversion is not necessary and low pressure drop is required, a horizontally sparged bubble column can be used (Joshi and Sharma, 1976). When complete gas phase conversion and hence long gas residence time is required, downflow bubble columns can be used. For example, phosgene decomposition is carried out advantageously in a downflow bubble column (Herbrechtsmeier and Steiner, 1980). In biotechnological processes low backmixing is desired, to achieve high substrate conversion. A continuous single-cell protein fermentation process is carried out in a sectionalized bubble column where low backmixing can be achieved (Schügerl et al., 1978). Vertically sparged bubble columns are most commonly used in the industry and hence, in this review, main emphasis is given to these types of columns. These columns are frequently used as absorbers, strippers and reactors in various chemical processes, such as coal liquefaction, fermentation, Fischer-Tropsch synthesis etc. A comprehensive list of processes in which bubble columns are used is given in Table 1.1.

Flow regime, bubble size distribution, and coalescence characteristics, gas holdup, interfacial mass transfer coefficients, gas-liquid interfacial area, dispersion coefficients and heat transfer characteristics are important design parameters for bubble column reactors. A thorough knowledge of these interdependent parameters is also necessary for a proper scaleup of these reactors. This review summarizes all the available pertinent literature for these design parameters and makes recommendations for the use of the available correlations.

FLOW REGIMES

In bubble column reactors the hydrodynamics, transport and mixing properties such as pressure drop, holdup of various phases,

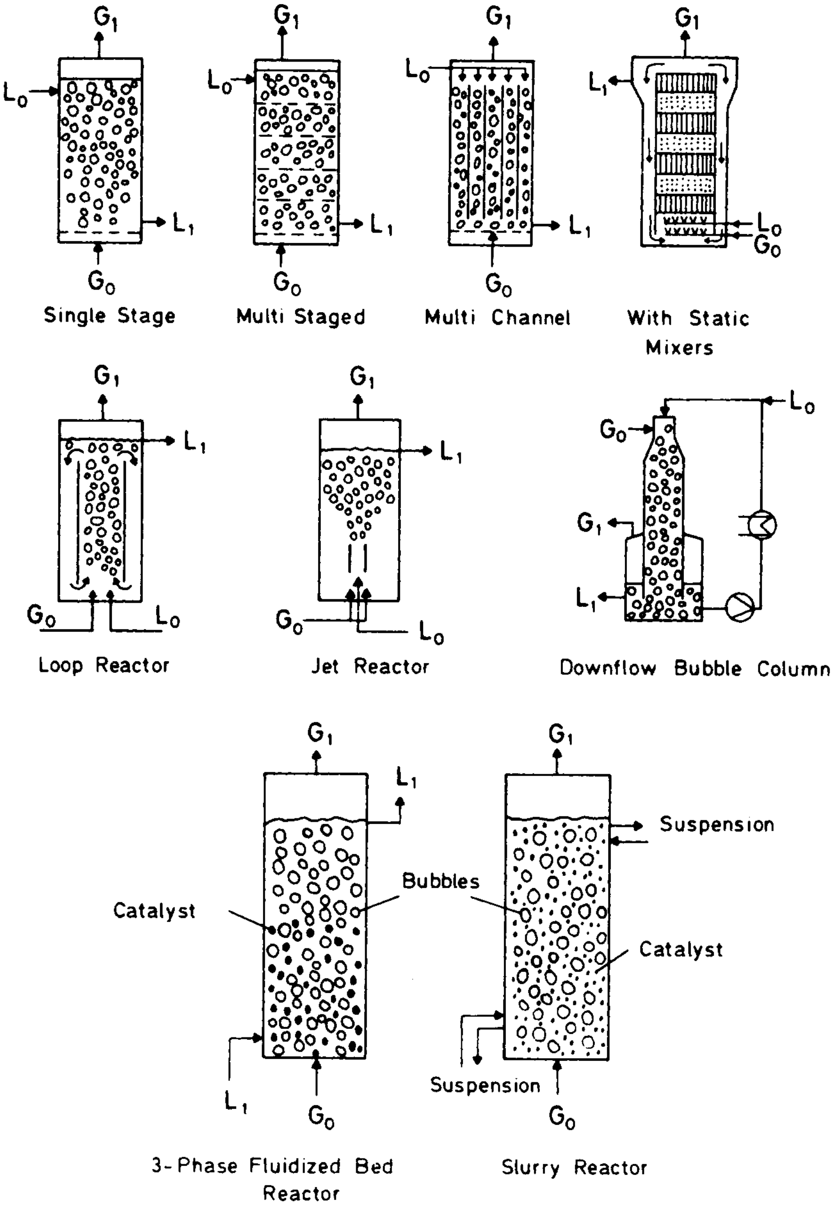

Figure 1.1. Bubble column reactors and modifications.

fluid-fluid interfacial areas, and interphase mass and heat transfer coefficients depend strongly on the prevailing flow regime. Many investigators (Govier and Aziz, 1972; Lockett and Kirkpatrick, 1975; Wallis, 1969; Kawagoe et al., 1976; Hills, 1976; Miller, 1980; etc.) have proposed different criteria to differentiate flow regimes. Wallis (1969) has characterized the upward movement of the bubble swarms into three separate flow regimes. These regimes occur in order of increasing gas flow rate.

1. Bubbly Flow or Quiescent Bubbling: This regime is characterized by almost uniformly sized bubbles with equal radial distribution. This regime occurs if the superficial gas velocity is less than 0.05 m/s (Fair, 1967) and the rise velocity of the bubbles lies between 0.18 and 0.30 m/s (Levich, 1962). However, these data should only be regarded as a guideline which are valid for aeration of water.

The theory of bubbly flow was developed by Lapidus and Elgin (1957), Richardson and Zaki (1954), and Wallis (1962). This approach gives satisfactory correlation of the liquid-gas bubble flow only if bubbles of equal size are uniformly distributed over the cross-sectional area. The theory fails if significant mass transfer occurs in the column (Deckwer et al., 1977b). Using stagnant bubble clouds in a flowing liquid, Lockett and Kirkpatrick (1975) showed that the bubble flow regime can be realized up to gas holdup of 66%, whereas in the usual arrangement with almost stagnant liquid the bubbly flow theory fails if the gas holdup is larger than about 15% (Deckwer et al., 1977b; Whalley et al., 1971).

2. Churn Turbulent Regime or Heterogeneous Regime: At higher gas velocities the homogeneous gas-in-liquid dispersion cannot be maintained and an unsteady flow pattern with channeling occurs. This heterogeneous flow regime is characterized by large bubbles moving with high rise velocities in the presence of small bubbles (Hills and Darton, 1976). The large bubbles take the form of spherical caps with a very mobile and flexible interface. These large bubbles can grow up to a diameter of about 0.15 m.

3. Slug Flow: In small diameter columns, at high gas flow rates, large bubbles are stabilized by the column wall leading to the formation of bubble slugs. Bubble slugs can be observed in columns of diameters up to 0.15 m (Hills, 1976; Miller, 1980).

Hills (1976) has extended the flow regime maps to high gas and liquid throughputs. Various flow regimes are schematically represented by Fig. 2.1. The dependence of flow regime on column diameter and gas velocity can be roughly estimated from Figure 2.2. The type of sparger used, physico-chemical properties of liquid, the liquid velocity can affect the transition between the flow regimes (Shah and Deckwer, 1981). For example: (i) slug flow can

TABLE 1.1. PRACTICAL EXAMPLES OF REACTIONS PROCESSED IN BUBBLE COLUMNS AND MODIFICATIONS

	System	Reference
A.	*Gas-Liquid Systems*	
1.	Absorption of CO_2 in Ammoniated Brine for the Manufacture of Soda Ash	Danckwerts and Sharma (1966), Kafarov et al. (1976)
2.	Absorption of CO_2 in Aqueous Buffer Solutions and Amines	Danckwerts and Sharma (1966), Danckwerts (1970)
3.	Absorption of Isobutylene, Butenes in Aqueous Solutions of H_2SO_4	Gehlawat and Sharma (1968), Kröper et al. (1969), Deckwer et al. (1977a), Deckwer (1977)
4.	Partial Oxidation of Ethylene to Acetaldehyde	Smidt et al. (1959), Smidt et al. (1962), Jira et al. (1976)
5.	Oxidation of Ethylene in Acetic Acid Solutions to Vinyl Acetate	Krekeler and Schmitz (1968)
6.	Oxidation of Acetaldehyde to Acetic Acid	Sittig (1967), Kostyak et al. (1962)
7.	Oxidation of Acetaldehyde to Acetic Anhydride	Yau et al. (1968)
8.	Acetic Acid Manufacture from Oxidation of Sec. Butanol	Sittig (1967)
9.	Oxidation of Butanes to Acetic Acid and Methyl Ethyl Ketone	Broiche et al. (1963), Broich (1964), Höfermann (1964), Saunby and Kiff (1976)
10.	Oxidation of Toluene to Benzoic Acid	Kaeding et al. (1965)
11.	Oxidation of Xylene to Phthalic Acid	Haase (1972)
12.	Oxidation of Cumene to Phenol and Acetone	Hattori et al. (1970), Hagberg and Krupa (1976)
13.	Oxidation of Ethylbenzene to Acetophenone	Sittig (1967)
14.	Wet Oxidation of Waste Water	Beyrich et al. (1979), Perkow et al. (1980)
15.	Alkylation of Phenols with Isobutylene Diluted with Inert Gas	Gehlawat and Sharma (1970)
16.	Carbonylation of Methanol to Acetic Acid	von Kutepow et al. (1965), Hjortkjaer and Jensen (1976)
17.	Hydroformylation of Olefins to Aldehydes and Alcohols	Oliver and Booth (1970), Weissermel and Arpe (1976)
18.	Oxychlorination of Ethylene to Dichloroethane	Albright (1967b), Friend et al. (1968), Rassaerts and Witzel (1974)
19.	Chlorination of Aliphatic and Aromatic Hydrocarbons	Hawkins (1965), Sittig (1967), van den Berg (1977), Rathjen (1975)
20.	Separation of Oily Water	Takahashi et al. (1979)
B.	*Gas-Liquid-Solid Systems*	
1.	Absorption of SO_2 in an Aqueous Slurry of Magnesium Oxide and Calcium Carbonate	Volpicelli and Massimilla (1970), Sada et al. (1977)
2.	Production of Aluminum Alkylates	Albright (1967d), Müller (1974)
3.	Wet Oxidation of Waste Sludge	Ploos van Amstel and Rietema (1970, 1973)
4.	Oxydesulfurization of Coal	Joshi et al. (1981)
5.	Coal Liquefaction	Wu and Storch (1968), Krönig (1977), Shah et al. (1978a), Frank and Knop (1979), Shah (1981)
6.	Polymerization of Olefins	Albright (1967a), Reichert (1977), Gates et al. (1979)
7.	Catalytic Desulfurization of Petroleum Fractions	Karolyi et al. (1963), Shah (1979)
8.	Upgrading of Coal Oils and Heavy Oil Fractions by Hydrogenations	Van Driesen and Stewart (1964), Ostergaard (1966)
9.	Hydrogenation of Unsaturated Fatty Acids	Albright (1967c), Hashimoto et al. (1971) van der Plank et al. (1972), Baltes et al. (1975)
10.	Hydrogenation of Benzene to Cyclohexane	Dufau et al. (1964), Bernard (1975)
11.	Hydrogenation of Adipic Acid Dinitrile to Hexamethylene Diamine	Weissermel and Arpe (1976)
12.	Hydrogenation of Nitroaromatics to Amines	Albright et al. (1967)
13.	Hydrogenation of Glucose to Sorbitol	Hofmann and Bill (1959), Haidegger et al. (1968), Brahme and Doraiswamy (1976)
14.	Hydrogenation of Anthraquinone to Anthrahydroquinone (in the production of hydrogen peroxide)	Williams and Lefeuvre (1956), The Industrial Chemist (1959)
15.	Hydrogenation of Ammonium Nitrates to Hydroxyl Amines	van Goolen (1976)
16.	Hydrogenation of α-nitrocaprolactum	van Dierendonck and Nelemans (1972)
17.	Oxidation of Cyclohexane to Mixture of Cyclohexanol and Cyclohexanone	Berezin et al. (1966), Bokelmann (1975)
18.	Oxidation of Cyclohexene to Adipic Acid	Berezin et al. (1966), Leitner (1974)
19.	Oxidations of n-parrafins to sec-alcohols	Kurata and Koshida (1978)
20.	Oxidation of Glucose to Gluconic Acid	de Wilt and van derBaan (1972)
21.	Methanol from Synthesis Gas	Sherwin and Frank (1976)
22.	Methanation of CO	Kölbel et al. (1965), Hammer (1968), Blum et al. (1975), Frank (1977)
23.	Fischer-Tropsch Synthesis	Kölbel and Ackermann (1956), Kölbel and Ralek (1977, 1980), Deckwer et al. (1980a, 1981a,b,c)
24.	Production of Hydrocarbons from CO and Steam	Kölbel and Engelhardt (1952), Kölbel et al. (1956)
25.	Production of Single Cell Protein	Rosenzweig and Ushio (1974), McLaren (1975), Moo-Young (1975), Schügerl et al. (1978), Sittig and Heine (1977), Sittig et al. (1978)
26.	Production of Primary and Secondary Metabolites	Smith and Greenshields (1974), König (1980)
27.	Production of Animal Cells	Katinger et al. (1979)
28.	Waste Water Treatment	Bayer (1977), Leistner et al. (1979), Zlokarnik (1979)

already occur at very low superficial gas velocity for highly viscous fluids (Schumpe, 1981); (ii) Anderson and Quinn (1970) have reported that minute traces of contaminants in the tap water can change the transition between flow regimes; (iii) porous spargers with mean pore sizes less than 150 μm generally produce bubble flow up to superficial gas velocities of about 0.05 to 0.08 m/s. While for perforated plates with orifice diameter larger than 1 mm, bubbly flow is possible only at low superficial gas velocities. At larger orifice diameters bubbly flow may not occur in case of pure liquids. Thus, the boundaries between the different flow regimes shown in Figure 2.2 are only approximate.

Flow regimes have been characterized by a few investigators either with the help of conductance probe or by visual observation (Javdani et al., 1977; Oshinowo and Charles, 1974; Barnea et al., 1980). Flow regime charts presented by these authors extend to higher gas and liquid flow rates which are usually uncommon in the application of bubble columns as chemical reactors and absorbers. Bach and Pilhofer (1978) have suggested that the churn-turbulent regime is most commonly encountered in industrial bubble columns. Knowledge of the transition from the bubble flow to the churn-turbulent and slug flow regimes is important because the achievable conversion depends strongly on the flow regime. Deckwer and Schumpe (1979) and Schumpe et al. (1979) demonstrated this effect by studying CO_2 absorption in alkali solution as a function of gas velocity for different column heights. As shown in Figure 2.3 conversion decreases very sharply from complete conversion as gas velocity is increased; a change which indicates a transition from bubble flow to churn turbulent flow. The con-

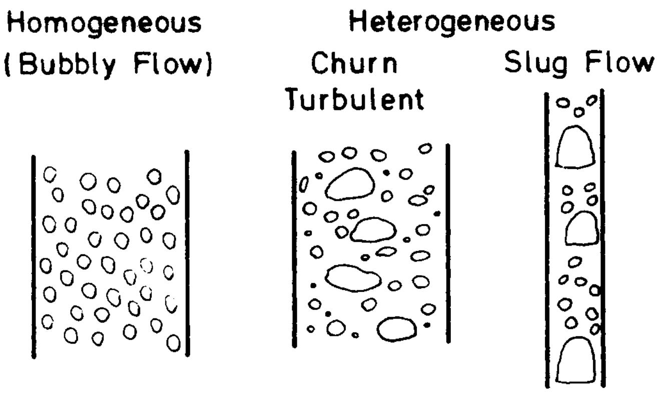

Figure 2.1. Flow regimes in bubble columns.

version drop can be explained by the results of Beinhauer (1971) and Kölbel et al. (1972) who measured fractional holdups and rise velocities of small ($d_{vs} < 0.005$ m) and large bubbles ($d_{vs} > 0.03$ m) in a 0.1 m diameter column for the air-water system. The fractional gas volume throughput of the large bubbles with short residence time in the column can be calculated from the data of Beinhauer (1971) and is given in Figure 2.4. For a superficial gas velocity of about 0.08 m/s, more than 60% of the gas flows through the reactor in the form of large bubbles with a rise velocity of about 0.85 m/s.

Wallis (1962) has suggested the use of flow regime charts based on the drift flux approach. The slip velocity U_s is defined as

$$U_s = \frac{u_G}{\epsilon_G} \pm \frac{u_L}{\epsilon_L} \tag{2.1}$$

Wallis (1962, 1969) defined the drift flux of gas as the volumetric flux of gas relative to a surface moving at average velocity, i.e.

$$j_{GL} = U_s\epsilon_G\,(1-\epsilon_G) = u_G\epsilon_L \pm u_L\epsilon_G \tag{2.2}$$

Lapidus and Elgin (1957) have suggested that the slip velocity is directly proportional to the terminal bubble rise velocity $u_{b\infty}$, i.e.

$$U_s = u_{b\infty}f_1(\epsilon_G) \tag{2.3}$$

Therefore, combining Eqs. 2.2 and 2.3,

$$j_{GL} = u_{b\infty}\epsilon_G(1-\epsilon_G)\,f_1(\epsilon_G) = u_{b\infty}f_2(\epsilon_G) \tag{2.4}$$

If the drift flux is plotted against the gas holdup, the change in the slope of the curve will indicate the transition from one regime to the other flow regime. Various expressions for drift flux are reported in the literature by Lockett and Kirkpatrick (1975). The updated list is given below.

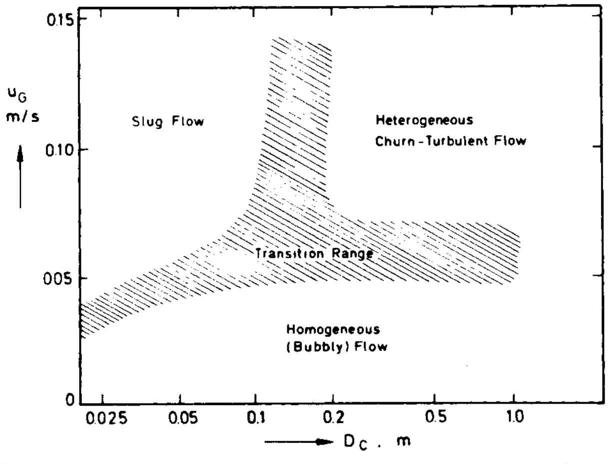

Figure 2.2. Approximate dependency of flow regime on gas velocity and column diameter (water and dilute aqueous solutions).

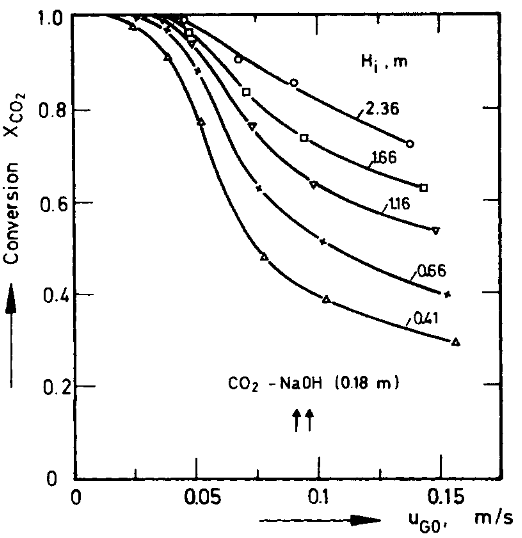

Figure 2.3. Conversion as function of gas velocity for chemical absorption of CO_2 (Schumpe et al., 1979).

$u_{b\infty}\epsilon_G(1-\epsilon_G)$ Turner (1966)

$u_{b\infty}\epsilon_G$ Davidson and Harrison (1966)

$u_{b\infty}\epsilon_G(1-\epsilon_G)^n$
where, $n = 2$ for small bubbles Griffith and Wallis (1961)
$= 0$ for large bubbles

$u_{b\infty}\epsilon_G(1-\epsilon_G)^{2.39}$ Richardson and Zaki (1954)

$u_{b\infty}\epsilon_G\dfrac{(1-\epsilon_G)^2}{(1-\epsilon_G{}^{5/3})}$ Marrucci (1965)

$u_{b\infty}\epsilon_G(1-\epsilon_G)^{2.39}\,(1+2.55\,\epsilon_G{}^3)$ Lockett and Kirkpatrick (1975)

$U_{b\infty}\epsilon_G(1-\epsilon_G)^m$ for three phase flows, Kara (1981)

$$m = -(1.951\times10^{-4}\left(\frac{D_C u_{SL}\rho_{SL}}{\mu_{SL}}\right) + 20.624\,\frac{\epsilon_S}{\epsilon_S+\epsilon_L} + 56.36\left(\frac{D_C u_t \rho_L}{\mu_L}\right) + 2.607)$$

Freedman and Davidson (1969) showed that for batch systems, Turner's (1966) expression described the measured data for air-water and air-electrolyte systems fairly well up to gas velocities of about 0.05 m/s. However, for holdup in commercial paraffin, the expression by Marucci (1965) described the data well. For gas holdup up to 30%, Richardson and Zaki's (1954) relation for drift flux is recommended since it fits most of the available literature data (Lockett and Kirkpatrick, 1975).

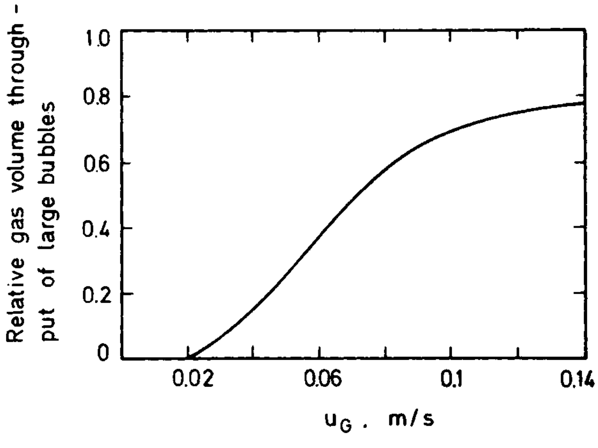

Figure 2.4. Fraction of volume throughput of large bubbles (calculated from Beinhauer, 1971).

In three phase flow systems, the effect of solid particles on the flow transition has been studied by some investigators (Darton and Harrison, 1975a,b; Javdani et al., 1976; Kara, 1981). Darton and Harrison proposed a trial and error procedure to calculate the value of gas and liquid holdup based on the drift flux theory, where the liquid holdup is empirically correlated to gas and liquid velocities and the ratio of clear liquid wake volume to bubble volume.

Zuber and Findley (1965) modified the drift flux theory for churn-turbulent regime. For cocurrent flow the drift flux equation can be written as

$$\frac{u_G}{\epsilon_G} = (u_G + u_L) + \frac{j_{GL}}{\epsilon_G} \tag{2.5}$$

or

$$\frac{\langle u_G \rangle}{\langle \epsilon_G \rangle} = \frac{\langle (u_G + u_L)\epsilon_G \rangle}{\langle \epsilon_G \rangle} + \frac{\langle j_{GL} \rangle}{\langle \epsilon_G \rangle} \tag{2.6}$$

where $\langle \quad \rangle$ bracket indicates the average value along the cross section. Zuber and Findley (1965) defined a distribution parameter

$$C_o = \frac{\langle \epsilon_G (u_G + u_L) \rangle}{\langle \epsilon_G \rangle \langle u_G + u_L \rangle} \tag{2.7}$$

which is a rough indication of non-uniform radial distribution. Equation 2.6 is rewritten as

$$\frac{\langle u_G \rangle}{\langle \epsilon_G \rangle} = C_o \langle u_G + u_L \rangle + \frac{\langle j_{GL} \rangle}{\langle \epsilon_G \rangle} \tag{2.8}$$

If the value of $\langle j_{GL} \rangle / \langle \epsilon_G \rangle$ is constant or very small compared to the value of $\langle u_G + u_L \rangle$, by plotting a graph of u_G/ϵ_G against $\langle u_G + u_L \rangle$, the value of C_o can be obtained. A change in the slope of the graph will indicate the bubbly flow regime. Zuber and Findley's theory can be modified for the three phase homogeneous slurries (Kara, 1981).

So far, only single stage bubble columns have been considered in this section. Insufficient data prevents any critical discussion about the flow regimes in novel reactors. Cocurrent downflow bubble columns can essentially only be operated in bubbly flow as the gas holdup values larger than 0.21 cannot be realized at atmospheric pressures. In bubble column loop reactors with high liquid circulation rates bubbly flow can be maintained up to higher gas velocities (≤ 0.1 m/s). Hence, larger amounts of gas can be processed through the reactor in bubbly flow (Weiland, 1978).

BUBBLE DYNAMICS

Bubble size, bubble rise velocity, bubble size distribution and liquid and bubble velocity profiles have a direct bearing on the performance of bubble columns.

Many methods are available to determine bubble sizes. Photographic techniques are used widely because of their simplicity (Kölbel et al., 1961; Akita and Yoshida, 1974; Burckhart and Deckwer, 1975; Schügerl et al., 1977; Quicker and Deckwer, 1981a). Other methods involve light scattering, light reflection (Calderbank et al., 1960) and depolarization (Rodionow and Patzelt, 1969). In the past decade, various optical and electrical probes have been developed to determine the bubble sizes (Todtenhaupt, 1971; Pilhofer and Miller, 1972; Burgess and Calderbank, 1975; Serizawa, et al., 1975; Calderbank and Pareira, 1977; Yamashita et al., 1979; Koide et al., 1979). Recently, a new dynamic bubble disengagement technique has been proposed to find out the bubble distribution (Sriram and Mann, 1977; Schumpe, 1981; Vermeer and Krishna, 1981). A review and comparison of the various probes is given by Buchholz et al. (1979, 1981) and Buchholz and Schügerl (1979a, 1979b). Though the original bubble size distributions obtained from various techniques differ markedly, the volume-to-surface mean bubble diameters d_{vs} (often called as Sauter mean diameter) evaluated consequently differ only slightly. This is confirmed by Yamashita et al. (1979); who have reported that the photographic results are in reasonable agreement with those obtained from the two-point electrical probe method. However, it should be pointed out that photographic, electrical, and optical probe techniques give reliable results only in the homogeneous or bubbly flow regime, e.g., at low gas velocities (say $u_G \leq 0.05$ m/s). In particular, the photographic technique is only limited to the bubbles in the vicinity of the column wall, but not the large bubbles which tend to rise in the central part of the column at high gas velocities and therefore sometimes predicts wrong values of mean bubble diameter.

On the basis of Kolmogoroff's isothropic turbulence theory, Calderbank (1967) proposed a number of equations for d_{vs} in stirred vessels. All the equations have the general structure,

$$d_{vs} = C \frac{\sigma^{0.6}}{(P'/V_D)^{0.4} \rho_L{}^{0.2}} \epsilon_G^n \left(\frac{\mu_G}{\mu_L}\right)^{0.25} \tag{3.1}$$

where the constant C and exponent n depend on the stirrer type and the kind of liquid phase. (P/V_D) represents the energy dissipation rate per unit volume of dispersion. If the gas is sparged by two-phase nozzles, correlation like Eq. 3.1 also apply to bubble columns (Nagel et al., 1972a, b, 1973, 1978, 1979; Unno and Inoue, 1980; Joshi, 1980; Kubie, 1981). In case the gas is sparged by single orifices, or perforated or sintered plates, the diameter of the generated bubbles is only slightly dependent on the gas velocity (Akita and Yoshida, 1974; Schügerl et al., 1977; Deckwer et al., 1978). Akita and Yoshida (1974) determined the bubble size distribution in bubble columns using a photographic technique. The gas was sparged through perforated plates and single orifices, and various liquids (water, aqueous and pure glycol, methanol, carbon tetrachloride) were used. On the basis of their data, Akita and Yoshida proposed the following correlation:

$$\frac{d_{vs}}{D_c} = 26 \left(\frac{D_c^2 g \rho_L}{\sigma}\right)^{-0.5} \left(\frac{g D_c^3}{\nu_L{}^2}\right)^{-0.12} \left(\frac{u_G}{\sqrt{g D_c}}\right)^{-0.12} \tag{3.2}$$

The correlation is based on data in columns up to 0.3 m in diameter and up to superficial gas velocities of about 0.07 m/s. Though Eq. 3.2 indicates that d_{vs} is proportional to $D_c^{-0.3}$; for $D_c > 0.3$ m, bubble diameter becomes independent of column diameter. Koide et al. (1979) reported bubble diameters in a 5.5 m diameter column and found larger sizes than those predicted from Eq. 3.2 and from a correlation derived earlier by Hughmark (1967). Koide et al. (1979) attribute this behavior to the use of relatively large nozzle diameter and insufficiently developed bubbly flow in axial and radial directions.

Akita and Yoshida (1974) found no effect of orifice diameter in a bulk region away from sparger. Obviously, a balance between coalescence and breakup rate controlled bubble sizes. This is also reported by Deckwer et al. (1978) who measured bubble sizes at 3.5 and 6.0 m above the sparger and found no significant differences in d_{vs} values.

The correlation of Akita and Yoshida can only be recommended for the systems studied by themselves and it seems particularly suited for single orifice spargers. For instance, bubble sizes in various hydrocarbons do not follow Eq. 3.2 (Quicker and Deckwer, 1981a,b). Bubble sizes in organic liquids were also reported by Hammer and Rähse (1973) and Hammer and Deckert (1978). Bubble sizes and their distributions for highly viscous Newtonian (aqueous glycerol) and non-Newtonian solutions (CMC (Carboxy Methyl Cellulose) and PA (PolyAcrylate)) were measured by Buchholz et al. (1978), Franz et al. (1980a,b), Nakanoh and Yoshida (1980), Schumpe (1981) and Schumpe and Deckwer (1981).

In contrast to Akita and Yoshida (1974), Kumar et al. (1976) and Bhavaraju et al. (1978) proposed correlations for bubble sizes which involve the orifice Reynolds number. The effect of the sparger is therefore still not clarified. Additionally, bubble diameter depends on the specific gas-liquid system and its properties with respect to coalescence. Coalescence is significantly influenced by the physical properties of the liquid. Calderbank (1967) and Vasalos et al. (1980) have reported high rates of bubble coalescence in highly viscous liquids. Bubble breakup is due to disturbances at the interface caused by external factors. Low viscosity liquid systems are observed to show significant bubble breakup. Schügerl et al. (1977) and Oels et al. (1978) used various liquid media for simulating fermentation broths and discussed their results with respect to the coalescence promoting and hindering properties of the liquid media and on the basis of the bubble size stability diagram developed by Berghmans (1973). For a given Bond number defined by

$$\mathrm{Bd} = \frac{\rho_L g d_b^2}{4\sigma}, \tag{3.3}$$

Berghmans' analysis gives a critical Weber number

$$\mathrm{We} = \frac{\rho_L u_{b\infty}^2 d_b}{2\sigma} \tag{3.4}$$

which divides the regions of stable and unstable bubble sizes as shown in Figure 3.1. The coalescence properties are mainly dependent on the added salts and the organic substances present. If a bubble is generated, the concentrations of an electrolyte at the interface and in the bulk liquid are initially equal. However, the ions have a tendency to move away from the interface. This results in an enrichment of the liquid phase, accompanied by an increase in the surface tension. Since the transport of ions in the bulk liquid requires some time, the coalescence hindering action is only pronounced at short residence times of the bubbles. Therefore large effects of added electrolytes on d_{vs} (and hence on interfacial area a and volumetric mass transfer coefficients) can be observed in stirred vessels and multistage columns where the life expectancy of individual bubbles is short. For instance, Zlokarnik (1978) reports a 700% increase in $k_L a$ in stirred aerated vessels. If the bubble residence time is as large as in single-stage tower fermenters, the influence of added electrolytes is much less pronounced. Therefore, Deckwer et al. (1978) did not find significant differences in bubble diameters for water and electrolytes in tall bubble columns. Under such conditions the analysis of Berghmans should apply, and indeed measured bubble diameters are located in the vicinity of the curve for the dynamic equilibrium bubble diameter (Schügerl et al., 1977) as shown in Figure 3.1.

The addition of alcohols lowers the surface tension if these substances have enough time to accumulate at the surface. As they orient themselves with their hydrophobic group towards the gas phase, the generated dipole layer suppresses the coalescence. The effect largely depends on the chain length of the alcohol. In addition, the gas distributor plays an important role. If sintered plates of high porosity and two-phase nozzles of injector or ejector type are applied as spargers, small bubbles are observed over the entire column. As shown in Figure 3.1, bubble sizes are in the stable region and considerably below the dynamic equilibrium curve. These considerations and conclusions of Schügerl et al. (1977) and Voigt and Schügerl (1979) present some useful guidelines for producing small bubble diameters in gas-liquid processes. Usually small bubbles are desirable as they yield large interfacial areas, which more than offsets the decrease in k_L with decreasing diameter.

The majority of bubble column reactors used in industry are operated at high pressures. Kölbel et al. (1961) studied the influence of pressure on the bubble size for air-water system. Up to pressures of 1.6 MPa they found no effect on the gas holdup and mean bubble diameter if the gas velocity is corrected to take into account the pressure in the column.

The mean rise velocity of the bubbles in a bubble swarm is equivalent to the interstitial gas velocity which follows from the superficial gas velocity

$$u_G^* = u_G / \epsilon_G \tag{3.5}$$

In the bubbly flow regime the rise velocity may vary from 0.03 to 0.2 m/s (Kölbel et al., 1961; Schügerl et al., 1977; Oels et al., 1978). If the flow is churn-turbulent, rise velocities are considerably higher (≥ 0.8 m/s) (Kölbel et al., 1972; Beinhauer, 1971). The transition from bubbly to churn-turbulent flow is usually accompanied by a sharp increase in u_G^*. The presence of surfactants results in decrease in the bubble rise velocities. This decrease is explained on the basis of surface tension gradient present on the bubble surface (Levich, 1962; Davis and Acrivos, 1966).

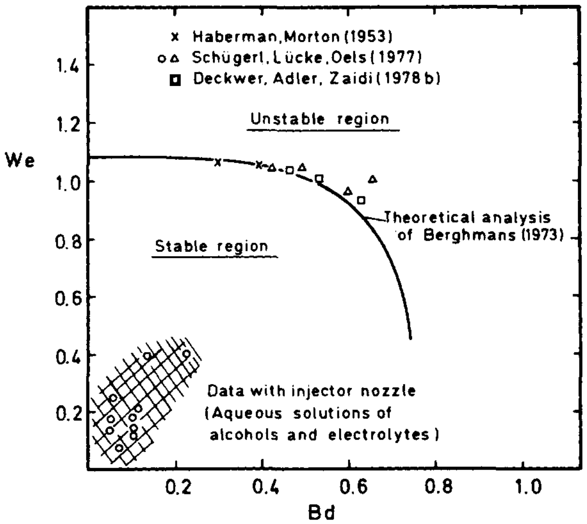

Figure 3.1. Application of Berghmans (1973) stability diagram of various aqueous solutions.

The terminal rise velocity of single bubbles ($u_{b\infty}$) is often used as a correlating parameter. This velocity can be calculated by the method of Clift et al. (1978).

$$u_{b\infty} = \frac{\mu_L}{\rho_L d_{vs}} M^{-0.149} (J - 0.857) \tag{3.6}$$

where

$$\mathrm{M} = \frac{g \mu_L^4 (\rho_L - \rho_G)}{\rho_L^2 \sigma^3}, \quad \mathrm{Re} = \frac{d_{vs} u_{b\infty} \rho_L}{\mu_L} \tag{3.7}$$

$$\mathrm{Eo} = \left(\frac{g(\rho_L - \rho_G) d_{vs}^2}{\sigma} \right) \tag{3.8}$$

$$J = 0.94\, \mathrm{H}^{0.747}\ (2 < \mathrm{H} \leq 59.3) \tag{3.9}$$

and

$$J = 3.42\, \mathrm{H}^{0.441}\ (\mathrm{H} > 59.3) \tag{3.10}$$

where

$$\mathrm{H} = \frac{4}{3} (\mathrm{Eo})\, \mathrm{M}^{-0.149} \left(\frac{\mu_L}{\mu_w} \right)^{-0.14} \tag{3.11}$$

for the range $\mathrm{M} < 10^{-3}$, $\mathrm{Eo} < 40$, $\mathrm{Re} > 0.1$. The equation covers an extensive range of data, and is therefore recommended for the calculation of terminal bubble rise velocity.

The radial nonuniformity in bubble columns can be partly explained with the help of liquid circulation rate. Different models have been proposed to calculate the liquid circulation rate and have been critically reviewed by Joshi and Shah (1980).

GAS HOLDUP

Gas holdup is one of the most important parameters characterizing the hydrodynamics of bubble columns. It can be defined as the percentage by volume of the gas in the two or three phase mixture in the column. It has two fold applications. On one hand, gas holdup in two phase systems gives the volume fraction of the phases present in the reactor and hence their resistance time. On the other hand, the gas holdup in conjunction with the knowledge of mean bubble diameter d_{vs}, allows the determination of interfacial area and thus leads to the mass transfer rates between the gas and liquid phase. Gas holdup depends mainly on the superficial gas velocity, and often is very sensitive to the physical properties of the liquid.

TABLE 2.1. HOLDUP CORRELATIONS IN LITERATURE (ALL DIMENSIONAL QUANTITIES ARE IN SI UNITS)

System	Range of Parameters	Correlation Proposed	Reference
1. Air-Water Air-Kerosene Air-Na_2SO_3 aq. soln. Air-Glycerol Air-Light Oil Air-$ZnCl_2$ aq. soln. ρ_L, kg/m^3: 780–1700 μ_L, Pa.s: 0.0009–0.0152 σ, N/m: 0.025–0.076	u_G, m/s: 0.004–0.45 D_C, m > 0.1	$\epsilon_G = \dfrac{1}{2 + (0.35/u_G)[\rho_L\sigma/72]^{1/3}}$	Hughmark (1967)
2. Air-Water	u_G, m/s: 0–0.3 u_L, m/s: 0–0.015 D_C, m: 0.066–0.214 H_t, m: 2.01–4.05	$\epsilon_G = \dfrac{2.51\,u_G}{\{0.78 + \beta u_G^{0.8}(1 - e^{\gamma})\}}$ $\beta = 4.5–3.5 - 2.548\,D_C^{1.8}$ $\gamma = 717\,u_G^{1.8}/\beta$	Kato and Nishiwaki (1972)
3. Air-Water	u_G, m/s: 0–0.26 u_L, m/s: 0.014–0.102	$\epsilon_L = 1.02\left(\dfrac{u_L^2\,\rho_G}{gD_C\rho_L}\right)^{-0.009}\left(\dfrac{u_G^2\,\rho_L}{gD_C\rho_G}\right)^{-0.036}$ $* \left(\dfrac{D_C u_G \rho_G}{\mu_G} \cdot \dfrac{D_C u_L \rho_L}{\mu_L}\right)^{-0.015}$	Kim et al. (1972)
Air-Water-Solids Air-Acetone aq. soln.-Solids Air-Sugar aq. soln.-Solids ρ_S, kg/m^3: 2300–2950 ρ_L, kg/m^3: 960–1170 μ_L, Pa·s: 0.001–0.07 σ, N/m: 0.0398–0.0738	d_p, mx10^{-3}: 1.0–6.0	$\epsilon_L = 1.504\left(\dfrac{u_L^2}{gd_p}\right)^{0.234}\left(\dfrac{u_G^2}{gd_p}\right)^{-0.086}$ $* \left(\dfrac{u_L^{2-n} d_p^n \rho_L}{v}\right)^{-0.082}\left(\dfrac{u_G v}{\sigma}\right)^{0.092}$ $v = g_c K\,8^{n-1}$ K = fluid consistency index n = fluid behavior index	Kim et al. (1975)
4. Air-Water Air-Glycol aq. soln. Air-Methanol O_2-Water He-Water CO_2-Water ρ_L, kg/m^3: 800–1600 μ_L, Pa·s: 0.00058–0.021 σ, N/m: 0.022–0.0742	u_G, m/s: 0.003–0.4 u_L, m/s: 0–0.044 D_C, m: 0.152–0.6 H_t, m: 1.26–3.5	$\dfrac{\epsilon_G}{(1 - \epsilon_G)^4} = C\left(\dfrac{gD_C^2\rho_L}{\sigma}\right)^{1/8}\left(\dfrac{gD_C^3}{\nu_L^2}\right)^{1/12}$ $* \left(\dfrac{u_G}{\sqrt{gD_C}}\right)$ C = 0.2 for pure liquids and non-electrolytes C = 0.25 for electrolytes	Akita and Yoshida (1973)
5. Air-Water Air-Methanol aq. soln. ρ_L, kg/m^3: 910–1200 μ_L, Pa·s: 0.0007–0.0138 σ, N/m: 0.0375–0.0748	u_G, m/s: 0.042–0.38 D_C, m: 0.1–0.19 H_t, m: 0.6–1.35	$\epsilon_G = 0.505\,u_G^{0.47}(0.072/\sigma)^{2/3}(0.001/\mu_L)^{0.05}$	Hikita and Kikukawa (1974)
6. Air-Different Liquids ρ_L, kg/m^3: 800–1600 μ_L, Pa·s: 0.00043–0.02 σ, N/m: 0.0214–0.0728	u_G, m/s: 0.01–0.08 D_C, m: 0.0756–0.61 H_t, m: 0.02–3.5	$\epsilon_G = 0.89\left(\dfrac{H_t}{D_C}\right)^{0.036(-15.7+\log K)}\left(\dfrac{d_b}{D_C}\right)^{0.3}$ $* \left(\dfrac{u_G}{d_b g}\right)^{2\ 0.025(2.6+\log K)} K^{0.047-0.05}$ $K = \rho_L\sigma^3/\mu_L{}_L^4 g$ $d_b = 0.003$ m	Gestrich and Rähse (1975)
7. Air-Water Air-Glycerol aq. soln. Air-Kerosene ρ_L, kg/m^3: 800–1100 μ_L, Pa·s: 0.0009–0.0115 σ, N/m: 0.0312–0.072	u_G, m/s: 0.0014–0.14 D_C, m: 0.05 and 0.1	$\epsilon_G = 0.728U - 0.485U^2 + 0.0975U^3$ $U = u_G\,[\rho_L^2/\sigma(\rho_L - \rho_G)g]^{1/4}$	Kumar et al. (1976)

TABLE 2.1. HOLDUP CORRELATIONS IN LITERATURE (ALL DIMENSIONAL QUANTITIES ARE IN SI UNITS)

8. Air-Ethanol-Solids Air-Glycerol aq. Soln.-Solids Air-Methanol-Solids Air-Water-Solids ρ_L, kg/m^3: 790–1210 μ_L, Pa·s: 0.001–0.062 σ, N/m: 0.0223–0.0728	u_G, m/s: 0.05–4.0 D_C, m: 0.05 and 0.1 d_p, m: 0.011–0.0287 H_t, m: 0.05–0.2	$\frac{\epsilon_G}{\{\epsilon_G(1-\epsilon_G)^2\}^{0.44}} = 0.5\left(\frac{D_C u_G^2 \rho_L}{\sigma}\right)^{0.11} * \left(\frac{u_G}{\sqrt{gD_C}}\right)^{0.22}$	Kito et al. (1976)
9. Air-Water-Solids	u_G, m/s: 0–0.173 u_L, m/s: 0–0.12 D_C, m: 0.076 and 0.152 H_t, m: 0.22–0.45	$\epsilon_G = (1.612 \pm 0.023) u_G^{0.720\pm0.028} * d_p^{0.168\pm0.061} D_C^{-0.125\pm0.088}$	Beovich and Watson (1978)
10. Air-Alcohols Air-Halogenated Hydrogens	u_G, m: 0–0.1 D_C, m: > 0.1 H_t, m: > 1.2	$\frac{\epsilon_G}{(1-\epsilon_G)} = 0.115\left[\frac{u_G^3}{\nu_L g(\rho_L-\rho_G)/\rho_L}\right]^{0.23}$	Bach and Pilhofer (1978)
11. Gas-Liquid	Ranges are not defined (semitheoretical equation)	$\frac{\epsilon_G}{(1-\epsilon_G)^4} = 0.14\, u_G \left(\frac{\rho_L^2}{\sigma(\rho_L-\rho_G)g}\right)^{1/4} * \left(\frac{\rho_L^2\sigma^3}{\mu_L^4(\rho_L-\rho_G)g}\right)^{1/24}$ $\left(\frac{\rho_L}{\rho_G}\right)^{5/72}\left(\frac{\rho_L}{\rho_L-\rho_G}\right)^{1/3}$	Mersmann (1978)
12. Different Gases (Air, H_2, CO_2, CH_4, C_3H_8, N_2)-Water Air-Organic Liquids Airj-Electrolyte Soln. ρ_L, kg/m^3: 790–1170 μ_L, Pa·s: 0.0009–0.0178 σ, N/m: 0229–0.0796 ρ_G, kg/m^3: 0.84–1.84	u_G, m/s: 0.042–0.38 D_C, m: 0.1 H_t, m: 0.65	$\epsilon_G = 0.672 f \left(\frac{u_G\mu_L}{\sigma}\right)^{0.578}\left(\frac{\mu_L{}^4 g}{\rho_L\sigma^3}\right)^{-0.131} * \left(\frac{\rho_G}{\rho_L}\right)^{0.062}\left(\frac{\mu_G}{\mu_L}\right)^{0.107}$ $f = 1.0$ for nonelectrolyte solutions $f = 10^{0.04141}\ I < 1.0$ kg ion/m^3 $f = 1.1 \quad I > 1.0$ kg ion/m^3 I = Ionic strength of the solution	Hikita et al. (1980)
13. Gas-Liquid	Theoretical (using stochastic approach)	$\frac{\epsilon_G}{(1-\epsilon_G{}^{1/3})} = \frac{u_G}{u_{b\infty}}$	Iordache and Nuntean (1981)
14. Downflow bubble Column			Friedel et al. (1980)
Air Ar H_2 CCl_2F_2 – Water CCl_4 Glycerol aq. soln. CMC	$148 \le I \le 336$ $0.003 \le \epsilon^* \le 0.24$ $184 \le \rho_L/\rho_G \le 5{,}340$ $37 \le \mu_L/\mu_G \le 2{,}220$ $0.055 \le \sigma \le 0.07$	$\epsilon_G = \left[1 + 0.0685\frac{(1-\epsilon^*)^{3.112}}{\epsilon^{*0.395}}\left(\frac{\rho_L}{\rho_G}\right)^{0.0346} * \left(\frac{\mu_L}{\mu_G}\right)^{0.254} \mathrm{Fr}^{0.36}\mathrm{We}^{0.543}\right]^{-1}$ $\epsilon^* = \frac{u_G}{u_G + u_L}$ $\mathrm{Fr} = I^2/(gD_C/(\rho_L\sigma)$ $\mathrm{We} = I^2 D_C/(\rho_L\sigma)$ I = Total mass flow density (kg/m^2·s)	

A column diameter greater than 0.15 m is sufficient to obtain the holdup values which are close to the ones obtained in larger diameter columns. Many investigators have obtained similar values of gas holdup for different diameter columns larger than 0.1 m (Miyauchi and Shyu, 1970; Kato et al., 1973; Akita and Yoshida, 1973; Hills, 1974; Hikita et al., 1980; etc.). Nicklin (1962) solved the equation of motion for bubbly flow and concluded that the mean slip velocity between bubble and liquid is approximately constant. Ueyama and Miyauchi (1979) proposed a hydrodynamic model by extending Nicklin's theory to the churn turbulent flow, and concluded that scale-up has very little effect on the gas holdup. Their analysis yields that the gas holdup in the churn turbulent flow is nearly proportional to $u_G^{0.5}$ and slightly decreases with an increase in the column diameter.

The dependence of the gas holdup on gas velocity is generally of the form,

$$\epsilon_G \alpha u_G^n \tag{4.1}$$

The value of n depends on the flow regime. For the bubbly flow regime value of n varies from 0.7 to 1.2 (Reith et al., 1968; Miyauchi and Shyu, 1970; Zlokarnik, 1971, Hammer and Rähse, 1973; Schügerl et al., 1977; Bach and Pilhofer, 1978; Botton et al., 1978; Deckwer et al., 1980a). In the churn turbulent or the transition regime, the effect of u_G is less pronounced and the exponent n takes values from 0.4 to 0.7. The log-log plots of gas holdup against gas velocity measured by various authors for air-water are illustrated in Figure 4.1. Only data for single orifice and multinozzle spargers are considered in this figure. Under such sparging conditions, the bubble flow regime at low gas velocities is often not very pronounced and the transition from the bubble flow to the churn-turbulent flow cannot be easily described. Although the data shown in Figure 4.1 are obtained in columns with diameters varying from

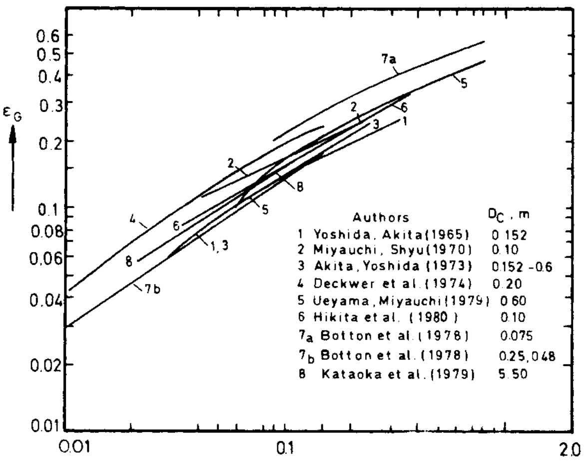

Figure 4.1. Experimental gas holdup as function of superficial gas velocity.

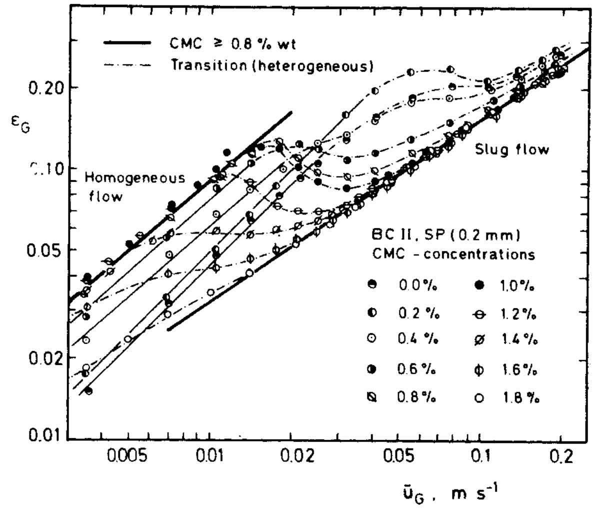

Figure 4.2. Gas holdup for CMC solutions (Schumpe et al., 1981).

0.075 to 5.5 m, the values of holdup match with each other indicating the relative unimportance of column diameter.

A large number of correlations for gas holdup have been proposed in the literature, however, the large scatter in the reported data does not allow a single correlation. Some of the important correlations have been reported in Table 2.1. The large scatter (Schügerl et al., 1977; Bach and Pilhofer, 1978) is mainly due to the extreme sensitivity of the holdup to the material system and to the trace impurities, which is not well understood. The easily available physical properties such as density, viscosity, and surface tension are not necessarily sufficient to explain the observed scatter. Only a few correlations are based on numerous experimental data (Akita and Yoshida, 1973; Hikita et al., 1980; Bach and Pilhofer, 1978; Mersmann, 1978). The correlation proposed by Mersmann (1978) is similar to the one obtained by Akita and Yoshida (1973). The equation proposed by Bach and Pilhofer (1978) describes the holdup data only for pure organic liquids and it does not apply to mixtures and electrolyte solutions. The gas holdup data even in mixtures of isomers like cis-decalin and trans-decalin differ from that in the pure liquids. Systematic measurements of the gas holdup in mixtures of organic compounds have been carried out by Bhaga et al. (1971), Hammer and Rähse (1973), Hammer and Deckert (1978) and Hammer (1979). These investigators observed that the experimental values cannot be correlated with the help of available literature correlations.

At higher temperatures (60 to 170°C) Quicker and Deckwer (1981a) determined the gas holdup for various types of spargers for hydrocarbon liquids (xylene, decalin, C_{10} to C_{14} paraffin mixtures, molten hard wax). The authors did not propose a new equation, but demonstrated that the correlations from literature failed to describe their data, though the physicochemical properties of the liquids are within the range of recommended for the application of these correlations.

Only the correlation of Hikita et al. (1980) considers physicochemical properties of the gas. As the small exponent of the gas properties in Table 4.1 indicates, the dependency is very small. However, Hikita et al. (1980) pointed out that the consideration of gas phase properties reduced the mean deviation of the correlated data from 15% to about 4%. One might suspect that at high pressures the gas holdup is significantly influenced by gas properties. But systematic studies have not been carried out so far. For the air-water system, the pressure dependency of the holdup was investigated by Kölbel et al. (1961) for the pressure range of 0.1 to 1.7 MPa. He found no effect of pressure, which is confirmed by Deckwer et al. (1980a), who measured the holdup for the pressure range of 0.4 to 1.1 MPa.

Jeffrey and Acrivos (1976) found that for particles as small as a few (<10) microns, suspensions behave macroscopically as non-Newtonian fluids. Therefore, gas holdup values involving the non-Newtonian liquids might be useful in analyzing the gas-slurry systems. Also, in bioreactors, the liquid shows non-Newtonian characteristics (Schumpe and Deckwer, 1981). Gas holdup data in non-Newtonian media have been reported by Nishikawa et al. (1977), Buchholz et al. (1978), Nakanoh and Yoshida (1980), and Schumpe et al. (1981). Gas holdup values higher than predicted by the correlation of Akita and Yoshida (1973) (Nakanoh and Yoshida, 1980) were obtained. At low gas velocities a bubbly flow regime can be realized with a sintered plate as shown in Figure 4.2. The holdup values do not depend on the CMC (carboxy methyl cellulose) concentration and hence viscosity. It can be seen from Figure 4.3 that, if perforated plates are used, slug flow exists for all the gas velocities. In the homogeneous flow regime the value of exponent n in Eq. 4.1 is found to be 0.85, while in the slug flow regime the value is 0.67. It should be noted that the effect of sparger is insignificant in the slug flow regime.

All the correlations given in Table 2.1 refer to the data obtained from single or multinozzle sparger with diameter of about 1 mm or larger. Hikita et al. (1980) compared their data with different correlations and they partly attributed the disagreement between their data to the differences in the design of gas spargers. They concluded that at low gas velocities, the single nozzle gas sparger gives lower values of gas holdup than a multinozzle or a porous plate sparger. Zahradnik and Kastanek (1979) carried out experiments with different hole diameter sieve plates and observed that the critical gas hole velocity corresponding to the onset of stable performance of a plate depends on the plate hole diameter. Nu-

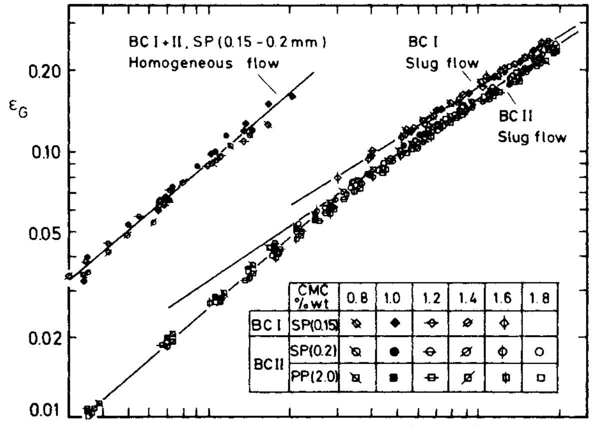

Figure 4.3. Correlation of gas holdup in CMC solutions for homogeneous flow and slug flow (Schumpe et al., 1981).

merous data for different kinds of spargers and various liquid media (synthetic fermentation media) have been reported by Schügerl et al. (1977), and Oels et al. (1978).

All these correlations predict average values of gas holdup over the whole reactor volume. Hills (1974) and Ueyama et al. (1980) have reported pronounced radial holdup profiles. Kobayashi et al. (1970) proposed an empirical correlation to evaluate the radial distribution. Also, axial variation in holdup can be observed at low gas velocities and at high conversions (Deckwer et al., 1978, 1980b).

The presence of solids does not affect the gas holdup significantly. Begovich and Watson (1978) combined the available 169 gas holdup points in three phase systems and proposed an empirical correlation. This equation is restricted to the batch liquid and solid systems. Recently Ying et al. (1980a) applied correlation by Akita and Yoshida (1973) to their data and concluded that the correlation by Akita and Yoshida is equally adequate for the three phase systems.

Liquid fluidized beds of small particles have been found to contract upon the introduction of gas (Stewart and Davidson, 1964; Ostergaard and Theisan, 1966; Kim et al., 1972), while the reverse trend occurs with large particles. The contraction can be explained by considering liquid wakes behind the gas bubbles. The liquid in the wake moves at a much faster rate than the continuous liquid phase. As a result, the average velocity of the bulk liquid phase decreases and the bed contracts causing an increase in the solid holdup. The large particles can cause a bubble breakup by virtue of their inertia and hence result in an expansion in fluidized beds. Bhatia et al. (1972), and Armstrong et al. (1976) studied the effect of solids wettability on the behavior of three phase fluidized beds of small and large particles respectively. They also have qualitatively described the role of wakes and bubbles in such beds. Kim et al. (1975) have reported the existence of two distinct types of three phase fluidization, which may be termed as a bubble coalescing and a bubble disintegrating. The former occurs when the particles are smaller than a critical size and the latter occurs when they are larger.

The axial solid concentration has been studied by Cova (1966), Imafuku et al. (1968), Farkas and Leblond (1969), Narayan et al. (1969), Yamanaka et al. (1970), and Govindrao (1975). Cova (1966) reported that high gas and liquid velocities and high viscosities tend to give more uniform solid distribution. Imafuku et al. (1968) observed that the critical gas velocity for complete suspension mostly depends on the liquid flow near the gas distributor, while Govindrao (1975) pointed out that particle diameter and bed volume have a strong influence on the axial distribution of solids. As a rule of thumb, in a cocurrent column, particles having sizes less than 100 μm diameter can form a pseudohomogeneous slurry, while particles greater than that will result in some axial solids distribution.

Kato et al. (1972a,b), Ying et al. (1980a), and Vasalos et al. (1980) investigated the effect of solids concentration and concluded that an increase in solids concentration generally decreases the gas holdup, but the effect becomes insignificant at high gas velocities (> 0.1 m/s).

In multistage columns Schügerl et al. (1977) observed higher values of gas holdup than in a single stage column. They also noted a strong effect of the distributor design in multistage columns for a noncoalescing medium. However, in air-water medium Zahradnik et al. (1974) and Zahradnik and Kastanek (1974) observed very little effect of the distributor design and the liquid velocity in multistage columns. Freedman and Davidson (1974) and Botton et al. (1978) carried out the experiments with the draught tubes, and observed insignificant effect of the draught tube on the holdup values. Weiland (1978) reported that in a bubble column with an external loop, the holdup values are comparable to the values in a single stage bubble column. As a rule of thumb it can be concluded that the effect of internals on the gas holdup is negligible, and any correlation which fairly represents the values of holdup in a single stage bubble column, can be used to calculate the values in the presence of internals.

The large number of correlations proposed for the holdup estimations indicates that an unique equation is not available. As a fair approximation, equations by Akita and Yoshida (1973) or Hikita et al. (1980) are recommended for less viscous and coalescing liquids ($\mu_L \leq 0.02$ Pa s). This equation can also be used for the three phase systems if the solid densities are close to water ($\approx$ 1,000–1,400 kg/m^3). For noncoalescing liquids (like surfactant solutions), or highly viscous liquids (non-Newtonian liquids) the following recommendation is given. As the gas holdup can be measured easily by various methods, it is better to carry out some measurements of the gas holdup in a lab scale column ($D_C \geq 0.15$ m). This will certainly give a better estimate for a specific design system than any available correlation.

GAS-LIQUID INTERFACIAL AREA

The gas-liquid interfacial area is an important design variable which depends on the geometry of the apparatus, the operating conditions and the physical properties of liquid media. The mass transfer rate, which is determined by the volumetric mass transfer coefficient k_La, can be easily influenced by varying interfacial area. Numerous techniques have been used to measure the interfacial area in gas-liquid dispersions. The methods fall into two main categories; namely, physical and chemical methods. The physical methods determine the size of the bubbles and these methods are mentioned in the bubble dynamics. From the gas holdup and volume-to-surface mean bubble diameter d_{vs}, the specific interfacial area can be calculated as

$$a = \frac{6\epsilon_G}{d_{vs}} \tag{5.1}$$

with

$$d_{vs} = \frac{\sum N_i d_{bi}^3}{\sum N_i d_{bi}^2} \tag{5.2}$$

d_{vs} can also be found from the correlations 3.1 and 3.2. Akita and Yoshida (1974) presented the following relation for the interfacial area per unit volume.

$$aD_C = \frac{1}{3}\left(\frac{gD_C{}^2\rho_L}{\sigma}\right)^{0.5}\left(\frac{gD_C^3}{\nu_L^2}\right)^{0.1}\epsilon_G^{1.13} \tag{5.3}$$

This correlation is based on the data with water, glycol solutions, methanol and carbon tetrachloride. Equation 5.3 shows a slight dependence on the column diameter, i.e., a is proportional to $D_C^{0.3}$. The influence of column diameter on the interfacial area vanishes at large column diameters, say $D_c \geq 0.3$ m. Akita and Yoshida's (1974) correlation is recommended as a conservative estimate of a at low gas velocities. Other correlations for interfacial areas of different systems can be found from the literature (Reith, 1968; Gestrich and Krauss, 1975; Kastanek, 1976; Schügerl et al., 1977).

If a chemical method is used to determine a, a suitable reaction is applied. The possible gas phase conversion X at reactor outlet is a function of the Stanton number

$$X = f\,(St_m) \tag{5.4}$$

The explicit expression of $f(St_m)$ depends on the reactor model applied and the reaction order. The Stanton number is defined by

$$St_m = K_m^*(aH_i/u_G) = K_m^*(6\tau/d_{vs}) \tag{5.5}$$

with the absorption-reaction parameter K_m^* defined as,

$$K_m^* = \left(\frac{2k_mD_i}{(m+1)\,H^{m+1}}\right)^{0.5} RTP^{(m-1)/2} \tag{5.6}$$

Table 3.1 gives a list of some convenient reaction systems and typical K_m^* values (Schumpe and Deckwer, 1980a). The widely applied reactions are sulfite oxidation and CO_2 absorption in alkali. Other suitable methods have been proposed in the literature, including the reactions in organic solvents (Sharma and Danckwerts, 1970; Bossier et al., 1973; Sridharan and Sharma, 1976; Dhanuka and Stepanek, 1980).

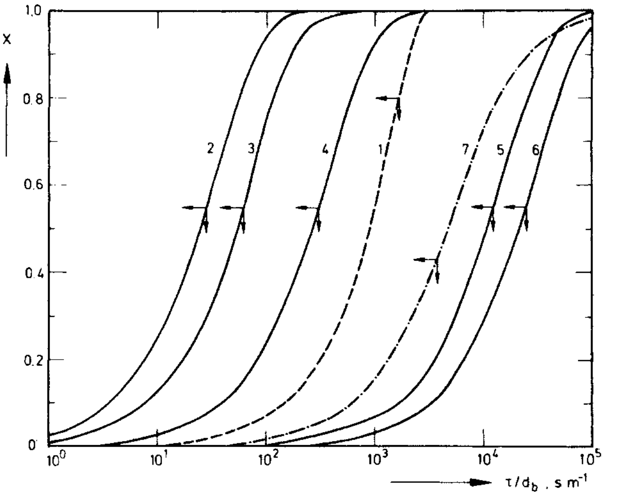

Figure 5.1. Conversion vs. τ/d_b for reactions listed in Table 3.1 (Schumpe and Deckwer, 1980a).

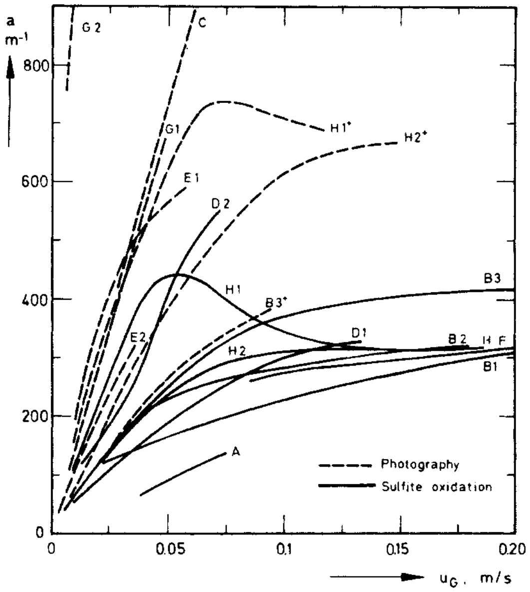

Figure 5.2. Comparison of interfacial areas from photography and sulfite oxidation (Studies listed in Table 3.2).

The interfacial area may deviate from the geometrical interfacial area and the discrepancy depends mostly on the reaction system and the conditions applied. Schumpe and Deckwer (1980a) have compared literature data on the effective interfacial areas from sulfite oxidation and CO_2 absorption into alkali and found considerable differences for the same contactor. The discrepancy is due to the use of an overall conversion which is incorrect, as the conversion depends nonlinearly on the interfacial area and in addition, the (τ/d_b) values of the bubbles usually show a broad distribution. The deviation of the chemical interfacial area from the geometrical area is larger when the distribution of τ/d_b is broader and when the Stanton numbers are higher. The error can be minimized if the measurements are carried out at low conversions. The dependence of the conversion on τ/d_b is plotted in Figure 5.1 and the reactions and their K_m^* values are given in Table 3.1. The arrows on each curve indicate the conversions and the values of τ/d_b or aH_t/u_G, respectively, which should not be exceeded to keep the deviation of the chemical area from geometrical one less than 15%. Schumpe and Deckwer (1980a) concluded that this gives a guideline for efficient experimentation which results in negligible differences between the two areas.

Interfacial areas determined from the physical and chemical methods may differ more than 100% (Voyer and Miller, 1968; Weisweiler and Rösch, 1978; Kürten and Zehner, 1978). Figure 5.2 shows experimental results obtained from the photographic and sulfite oxidation methods. The studies considered are summarized in Table 3.2. With the exception of the three points of Reith and Beek (1978) measured by photographic method (Curve $B3^+$), Figure 5.2 clearly indicates that the results of photographic and sulfite oxidation methods show large variations, particularly at high gas velocities. For the homogeneous bubbly flow Schumpe and Deckwer (1980b) have found

$$a_{\text{photo}} = 1.35\, a_{\text{sulfite}} \tag{5.7}$$

For the churn-turbulent flow the difference is even larger. This is a further proof that the photographic method is not very reliable. Thus, for the measurement of an interfacial area, use of a chemical method with low conversion is recommended. Sulfite oxidation is particularly recommended as an appropriate method.

Nagel and Kürten (1976) and Nagel et al. (1979) derived the following expression for the specific interfacial area based on the Kolmogoroff's theory of isotropic turbulence.

$$a = K\left(\frac{e'}{v_R}\right)^{0.4} \epsilon_G^n \tag{5.8}$$

This equation applies if the conditions of isotropic turbulence and a spatially uniform energy dissipation are fulfilled. Nagel et al. (1972a, b, 1973, 1978, 1979) measured interfacial areas with the sulfite oxidation method in various gas-liquid contactors and found that in practice, the exponent of the energy dissipation density varies between 0.4 and 1.0. Nagel et al. (1973, 1978) have presented the correlation for interfacial area in the graphical form which can be used in the design and scaleup of bubble column and other gas-liquid reactors. Oels et al. (1978) have modified the correlation for other systems and various types of gas spargers. The data and correlations of Oels et al. (1978) are based on the photographic method.

Recently, Schumpe and Deckwer (1981) used sulfite oxidation to measure the specific interfacial area in carboxy methyl cellulose solutions (0.5 to 1.6% wt.) in a 0.14 m diameter reactor. At gas velocities above 0.02 m/s the gas rises as slugs through the column. In this flow regime, the measured interfacial area can be described with striking agreement by

$$a = 48.7\,(u_G/\mu_{\text{eff}})^{0.51} \tag{5.9}$$

The effective viscosity μ_{eff} of the liquid phase in the aerated bubble column was obtained from the measured shear rate against shear

TABLE 3.1. ABSORPTION-REACTION SYSTEMS FOR THE INTERFACIAL AREA DETERMINATION

No.	Absorption-Reaction System	m	K_m^* m/s × 10^2	Reference
1	O_2 (air), 0.072 M $Na_2S_2O_4$, 0.0975 M NaOH	0	5.0×10^{-3}	Jhaveri and Sharma, 1968
2	CO_2, 2 M MEA, H_2O		0.496	Sridharan and Sharma, 1976
3	CO_2, 0.95 M NaOH		0.0223	Danckwerts and Sharma, 1966
4	CO_2, 2 M DEA, diethylene glycol	1	4.47×10^{-2}	Sridharan and Shrma, 1976
5	Isobutene, H_2SO_4 (50% wt), 2 M t-butanol		1.09×10^{-3}	Allenbach et al., 1977
6	O_2, 0.392 M CuCl, 5 M HCl		5.65×10^{-4}	Jhaveri and Sharma, 1967
7	O_2(air), 0.8 M Na_2SO_3, pH = 8, c(CO^{2+}) = 1×10^{-3} M	2	6.52×10^{-3}	Reith, 1968

TABLE 3.2. INVESTIGATIONS ON INTERFACIAL AREAS IN BUBBLE COLUMN CONSIDERED IN FIGURE 5.2 (SULFITE OXIDATION AND PHOTOGRAPHY OF GAS DISPERSION IN SULFATE SOLUTION)

Symbol		Bubble Column		Sparger Diameter and Number of Holes	Concentration of Na_2SO_3/Na_2SO_4 mol/l	Reference
Chem.	Photogr.	D_c, m	H_i, m			
A		0.153	1.3	PP*, 1.6 mm, 27	0.8	Carleton et al., 1967
B1		0.140	2.0–3.4			
B2		0.290	2.7	PP, 2 mm, 1 hole/cm^2	0.8	Reith and Beek, 1968
B3	B3$^+$	0.290	1.6			
	C	0.1–0.3	0.5–3.3	Ring, Cross	0.8	van Dierendonk et al., 1968
D1		0.10	1.0–1.4	PP, 1 mm, 59	0.5	Burkel, 1974
D2		0.10	1.0–1.6	SP*, 0.04/0.1/0.12 mm		
	E1	0.15	4.4	SP, 0.15 mm	0.11	Burckhart and Deckwer, 1975
	E2	0.20	7.2	Cross, 1 mm, 56	0.35	
F		0.14	3.3	PP, 2 mm, 84	0.8	Cornelius et al., 1977
	G1	0.14	4.0	PP, 0.5 mm, 180	0.8	Oels et al., 1978
	G2	0.14	4.0	SP, 0.018 mm		
H1	H1$^+$	0.102	2.4	SP, 0.15 mm	0.8	Schumpe and Deckwer, 1980a,b
H2	H2$^+$	0.14	2.7	PP, 0.5 mm, 421		

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* PP, perforated plate; SP, sintered plate.

stress curve by using the correlation of Nishikawa et al. (1977).

Sharma and Mashelkar (1970) observed that the effective interfacial area based on the dispersion volume always shows an improvement over unpacked bubble column at the same superficial gas velocity. Wang and Fan (1978) have indicated that for the bubble columns packed with motionless mixers even the effective interfacial area based on column volume is higher than that in the unpacked column. Juvekar and Sharma (1973) observed that the interfacial area in the presence of fine solid particles is a weaker function of gas velocity than the one in absence of solids.

Akita and Yoshida's correlation can be used for the estimation of conservative values of a. It applies particularly for the less effective gas spargers. In general, the relation given by Nagel et al. (1979) (Eq. 5.8) can be recommended. It has been proved to be valid for bubble columns aerated by two phase nozzle spargers and other kinds of gas-liquid contactors. For non-Newtonian highly viscous media Eq. 5.9 of Schumpe and Deckwer (1981) applies.

MASS TRANSFER COEFFICIENTS

Volumetric Gas-Liquid Mass Transfer Coefficients

Several studies (Mashelkar, 1970; Deckwer et al., 1974; Urza and Jackson, 1975; Burckhart, 1976; Buckhart and Deckwer, 1976; Gestrich et al., 1976; MacLean et al., 1977; Schügerl et al., 1977; Shioya and Dunn, 1978; Alvarez-Cuenca et al., 1980; Mangartz and Pilhofer, 1981; Deckwer et al., 1981d) indicate that the knowledge of the residence time distribution of the phases is necessary to determine the volumetric mass transfer coefficient, k_La. The assumption of complete mixing is only justified in large diameter columns, say $D_C \geq 1.0$ m. In tall and small diameter bubble columns the determination of k_La should be based on the concentration profiles measured at cocurrent or countercurrent flow along the column and evaluated by means of the axial dispersion model (Deckwer et al., 1974; Schügerl et al., 1977; Mangartz and Pilhofer, 1981). However, evaluation of the liquid phase concentration profiles can only be used in the presence of a large concentration difference along the reactor. This can be achieved by moderate gas velocities (usually $u_G < 0.1$ m/s) or high liquid flow rates. If the driving concentration difference becomes too small, i.e., the liquid is saturated to more than 90%, small errors in the concentration measurements may lead to unusually high k_La values. (See, for instance, Voigt and Schügerl, 1979.)

Volumetric mass transfer coefficients depend on the gas velocity, the sparger design and are sensitive to the physico-chemical properties, particularly, those which promote or prevent coalescence. In addition, the column diameter has some influence if it is small, say, $D_C \leq 0.15$ m. Furthermore, the k_La values may vary spatially. Sometimes larger k_La values are observed in the immediate vicinity of the sparger than in the bulk region (Deckwer et al., 1978, 1980b; Luttmann, 1980; Buchholz et al., 1980). Recently Alvarez-Cuenca and Nerenberg (1981) have introduced a two zone model to explain this phenomenon. Kastanek (1977) used Higbie's penetration theory, and by substituting the contact time by an expression derived from Kolmogoroff's theory of isotropic turbulence, he arrived at the following equation for k_La.

$$k_La \;\alpha\; \frac{u_G(u_G + C)^{13/20}}{2u_G + C} \qquad (6\text{-a.}1)$$

where C is a constant. Deckwer et al. (1974) proposed an empirical correlation of type

$$k_La = b\, u_G{}^n \qquad (6\text{-a.}2)$$

Table 4-a.1 summarizes measurements of k_La correlated by Eq. 6-a.2. The data have been obtained for tap water and salt solutions from several bubble columns equipped with sintered plates and nozzles of 1 mm diameter as gas distributors (Deckwer et al., 1974; Buckhart and Deckwer, 1976; Buckhart, 1976). Recently, Herbrechtsmeier and Steiner (1978) measured k_La values in a downflow bubble column and found a good agreement with the data reported by Deckwer et al. (1974). In agreement with Table 4-2.1, Kastanek et al. (1977) found n varying from 0.78 to 0.82, and being

TABLE 4-a.1. CORRELATION FOR VOLUMETRIC MASS TRANSFER COEFFICIENTS OF THE TYPE $k_La = bu_G{}^n$ FOR OXYGEN TRANSFER IN WATER AND ELECTROLYTE SOLUTIONS (k_La in s^{-1}, u_G in m/s)
(Data from Deckwer et al., 1974; Burckhart and Deckwer, 1976; Burckhart, 1976)

	Liquid Phase	Number of Data Points	b	n	Mean Error, %	Range of u_G, m/s
Bubble columns with cross of nozzles (1 mm)	Tap Water	66	0.467	0.82	17.7	0.002–0.08
D_C = 0.2 m, H_t = 2–7.2 m absorption, desorption	Salt Solutions (NaCl, Na_2SO_4)	49	0.460	0.79	7.5	0.004–0.08
Bubble columns with sintered plates (150 m)	Tap Water	61	1.174	0.82	16.6	0.003–0.08
D_C = 0.1–0.15 m, H_t = 2.5–4.4 m absorption, desorption	Salt Solutions (NaCl, Na_2SO_4)	51	1.445	0.78	13.9	0.004–0.04

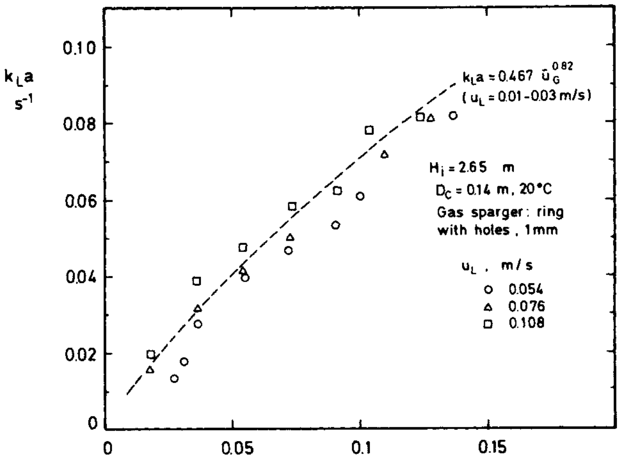

Figure 6-a.1. Effect of liquid velocity on k_La.

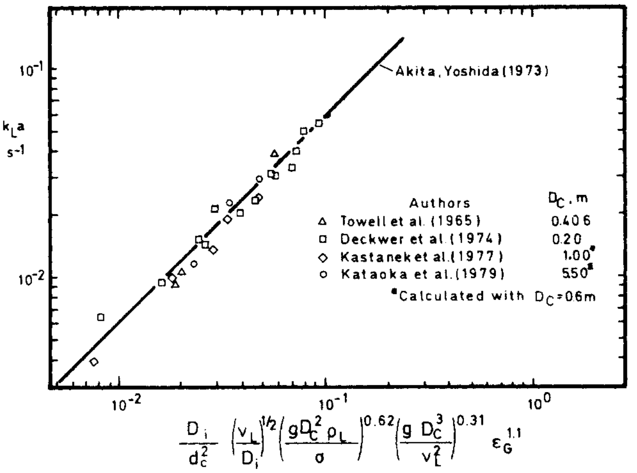

Figure 6-a.2. Correlation of Akita and Yoshida (1974)—comparison with data of other authors.

unaffected by the sparger. However, as evident from Table 4-2.1, the constant b is strongly affected by the sparger and the liquid medium. Porous distributors like sintered plates, elastic rubber plates (Rice et al., 1980; Deckwer et al., 1981d) and nozzle spargers of injector and ejector type (with gas and liquid flow) give higher k_La values than single and multinozzle spargers.

Comprehensive k_La data for oxygen mass transfer in the aqueous solutions of alcohols and glucose, with and without the presence of inorganic electrolytes in bubble columns equipped with various types of spargers are presented by Schügerl et al. (1977); Oels et al. (1978), and Voigt and Schügerl (1979). Though no correlations are developed, the experimental findings are explained by means of coalescence promoting and hindering properties of the liquid medium.

Some authors (Voyer and Miller, 1968; Alvarez-Cuenca et al., 1980) have reported the dependence of k_La on the liquid flow rate. However, it has been shown by Burckhart and Deckwer (1976) that this effect results from using the NTU method for the data evaluation. This method assumes plug flow for both phases in a bubble column. Figure 6-a.1 shows k_La data measured for the liquid velocities of 0.054, 0.076 and 0.108 m/s. These data indicate that an influence of u_L on k_La can be neglected. Also plotted in this figure is the first correlation of Table 6-a.1. A good agreement is found with this correlation which is based on the data measured at the liquid velocities between 0.007 and to 0.03 m/s.

Correlations for the volumetric mass transfer coefficients are summarized in Table 4-a.2. Figure 6-a.2 demonstrates the goodness of Akita and Yoshida's (1973) correlation in predicting k_La data measured in bubble columns of various sizes.

In the large diameter reactors the k_La does not depend on the column diameter. Therefore, the calculations for the 1 m and 5.5 m columns were done with $D_C = 0.6$ m as proposed by Kataoka et al. (1979). It has to be pointed out that the correlation of Akita and Yoshida (1973) applies well to the studies where gas is sparged by less effective spargers, i.e., either single or multiorifice distributors. The Akita-Yoshida correlation can therefore be recommended for a conservative estimation of k_La. Only Mangartz and Pilhofer (1981) report even lower k_La values, i.e., about 50% of those predicted from Akita and Yoshida's correlation. If gas spargers like porous plates and two component nozzles (injector and ejector) are used, k_La values, considerably higher than those calculated from the correlation of Akita and Yoshida can be obtained (Deckwer et al., 1974; Schügerl et al., 1977; Zlokarnik, 1979). However, no correlation has been established to describe these comprehensive data.

Oxygen mass transfer in highly viscous Newtonian and non-Newtonian liquids in bubble columns was measured by Buchholz et al. (1978), Voigt et al. (1980), Hecht et al. (1980), Nakanoh and Yoshida (1980) and Deckwer et al. (1981d). For gas velocities less than 0.1 m/s, Nakanoh and Yoshida (1980) have correlated all their data by the correlation given in Table 6-a.2. The apparent viscosity to be applied in this correlation was obtained from the viscosity against shear rate curve, and the shear rate $\dot{\gamma}$ was calculated from

$$\dot{\gamma} = 5000\, u_G \tag{6-a.3}$$

where $\dot{\gamma}$ in s^{-1} and u_G in m/s. Equation 6-a.3 was proposed by Nishikawa et al. (1977) from the heat transfer measurements in bubble columns.

Volumetric mass transfer coefficients in CMC solutions are also reported by Buchholz et al. (1978) for a single stage bubble column (0.14 m ID by 3.9 m height) and by Voight et al. (1980) for a multistage bubble column. k_La data for PAA (Poly Acryl Amide) solutions have been obtained by Hecht et al. (1980). The results with the CMC solutions were correlated by Henzler (1980). However, Henzler's correlation predicts an unusually low dependency on u_G, i.e., k_La is proportional to $u_G^{0.1}$ and the predictions deviate largely from Nakanoh and Yoshida's correlation. Therefore oxygen mass transfer in CMC solutions was remeasured in a 0.14 m diameter column by Deckwer et al. (1981d). The dependency of k_La on the gas throughput is complex, but for the most important flow regime, namely slug flow, which is established for all gas spargers if $u_G \geq 0.2$ m/s, all the measured data can be described by the fifth correlation given in Table 4-a.2. This equation gives lower values than the Nakanoh-Yoshida correlation but it describes k_La data measured in non-Newtonian fermentation broth (penicillin chrysogenium) with striking agreement. It is therefore believed that the correlation of Deckwer et al. (1981d) gives reliable k_La data for highly viscous non-Newtonian solutions (like CMC).

In the three phase bubble column reactors, k_La can be affected by the presence of solids. Investigations of various authors (Voyer and Miller, 1968; Slesser et al., 1968; Sharma and Mashelkar, 1968; Kato et al., 1973; Juvekar and Sharma, 1973; Sittig, 1977; Zlokarnik, 1979) indicate that the degree of influence of suspended particles on k_La depends on the particle concentration, the particle size, the liquid-solid density difference, the geometrical sizes and the operating conditions of the reactor (i.e., gas and liquid velocities). Some of the possible effects are demonstrated in Figures 6-a.3 and 6-a.4 for the aeration of a suspension of glass beads (300 μm) in water (Nguyen-Tien and Deckwer, 1981). At high liquid velocities (u_L = 0.093 m/s) and low gas velocities, the k_La values are slightly higher than those without the presence of solids. Such a small increase in k_La was also reported by Sittig (1977) for the solids (various plastic particles) concentrations below 15% wt. and particle sizes between 50 and 200 μm. With rising gas velocities and decreasing liquid velocities the particle distribution becomes increasingly nonuniform and the k_La values are lower than the ones without the solids. Obviously, the influence of the solid concen-

TABLE 4-a.2. REPORTED STUDIES FOR k_La

	System	Range of Parameters	Correlation Proposed	Reference
1.	Water-Air	$u_G\left(\frac{m}{s}\right)$: 0.003–0.4		Akita and Yoshida (1973)
	Glycol-Air	$u_L\left(\frac{m}{s}\right)$: 0.0–0.44	$\frac{k_LaD_C^2}{D_i} = 0.6\left(\frac{\nu_L}{D_i}\right)^{0.5}\left(\frac{gD_C^2\rho_L}{\sigma}\right)^{0.62}\left(\frac{gD_C^3}{\nu_L^2}\right)^{0.31}$	
	Methanol-Air			
	Glycol aq. soln.-Air	D_C (m): 0.152–0.6	$*\epsilon_G{}^{1.1}$	
	Methanol aq. soln.-Air	H_i (m): 0.126–0.35		
	Water-O_2	ρ_L (kg/m^3): 800–1600		
	Water-He	μ_L (Pa·s): 0.00058–0.021		
	Water-CO_2	σ (N/m): 0.022–0.0742		
2.	Gas-Liquid	Theoretical Equation	$k_La = 3.31\frac{D_i\epsilon_G}{d_b^2}\left(\frac{\mu_L}{\rho_L \cdot D_i}\right)^{1/3}$	Fair (1967)
		$*\left(\frac{d_b\rho_Lu_G}{\mu_L\epsilon_G}\right)^{1/2}$		
3.	Water-Air			
	Sucrose aq. soln.-Air	$u_G\left(\frac{m}{s}\right)$: < 0.1	$\frac{k_LaD_C^2}{D_i} = 0.09\left(\frac{\nu_{\text{eff}}}{D_i}\right)^{0.5}\left(\frac{gD_C^2\rho_L}{\sigma}\right)^{0.75}$	
	CMC aq. soln.-Air	μ_L(Pa·s): 0.005–0.06	$*\left(\frac{gD_C^3}{\nu_{\text{eff}}{}^2}\right)^{0.39}*\left(\frac{u_G}{gD_C}\right)$	Nakanoh and Yoshida (1980)
	Sodium polyacrylate aq. soln.,-Air	ρ_L(Kg/m^3): 995–1230 $C = 0$ for unelastic liquids $C = 0.133$ for elastic liquids $m = 0.55$ λ = characteristic relaxation time	$*\left(1 + C\left(\frac{u_{b\infty}\lambda}{d_{vs}}\right)\right)^{m-1}$	
4.	Water-Air			
	Water-O_2	$u_G\left(\frac{m}{s}\right)$: 0.042–0.38	$k_La = \frac{14.9gf}{u_G}\left(\frac{u_G\mu_L}{\sigma_L}\right)^{1.76}\left(\frac{\mu_L{}^4g}{\rho_L\sigma^3}\right)^{-0.248}$	Hikita et al. (1981)
	Water-H_2 Water-CH_4	H_i(m): 0.13–0.22 D_C(m): 0.10 0.19	$*\left(\frac{\mu_G}{\mu_L}\right)^{0.243}*\left(\frac{\mu_L}{\rho_LD_i}\right)^{-0.604}$	
	Water-CO_2 Sucrose solutions-Air n-butanol-air	ρ_L (Kg/m^3): 998–1230		
	Methanol solutions-Air	μ_L (Pa·s): 0.0008–0.011	$f = 1.0$ for nonelectrolytes	
	Electrolyte Solutions-Air	σ (N/m): 0.025–0.082	$f = 10^{0.068I}\ I < 1.0\,\frac{\text{Kg ion}}{\text{m}^3}$	
		D_i (m^2/s): 4.6–26.0	$f = 1.114 * 10^{0.021}\ I > 1.0\,\frac{\text{Kg ion}}{\text{m}^3}$	
5.	Aqueous CMC Solutions (1.0–2.0 wt %)	$u_G\left(\frac{m}{s}\right)$: 0.08		
		D_C (m): 0.14	$k_La = 0.00315\,u_G^{0.59}\mu_{\text{eff}}{}^{-0.84}$	Deckwer et al. (1981d)
		H_i (m): 2.6		

tration on k_La depends largely on the liquid and gas velocities. At high u_L (0.093 and 0.076 m/s in Figure 6-a.3) a clear dependency on solid concentration cannot be recognized even for high gas velocities. However, at a liquid velocity of 0.054 m/s (Figure 6-a.4) the decrease in k_La (in relation to those values without solids) depends on particle concentration if $u_G > 0.05$ m/s. Kato et al. (1973) have also shown that for higher solid concentration a steep decrease in k_La is found which is caused by a decrease in a. Recently, it has been shown by Dhanuka and Stepanek (1980) that with an increase in particle size, k_La decreases because of a decrease in a. For the typical operating conditions prevailing in the Fischer-Tropsch synthesis in slurry phase, Zaidi et al. (1979) and Deckwer et al. (1980a) have shown that the presence of solid particles (diameter less than 50 μm, concentration of solids < 16% wt) gives a negligible effect on k_La.

Weiland (1978) investigated the mass transfer in bubble column with an external loop. He reported an agreement of k_La data for water-O_2 with the data reported by Yoshida and Akita (1965) and Lin et al. (1976). Weiland (1978) observed that k_La data for aqueous salt solutions are larger by 50 to 100% compared to water.

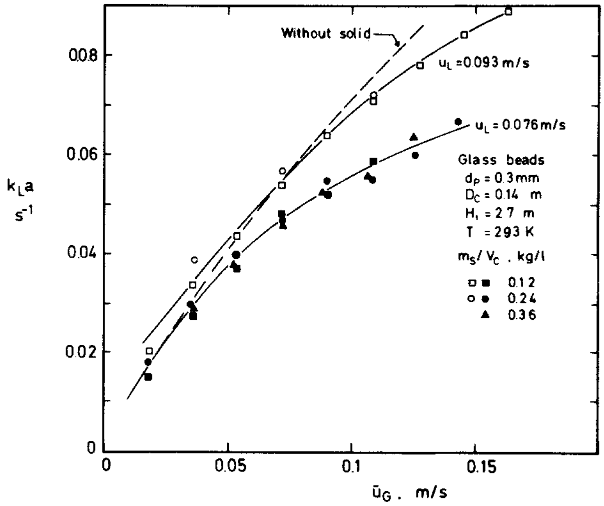

Figure 6-a.3. Effect of solids on volumetric mass transfer coefficient (Nguyen-Tien and Deckwer, 1981).

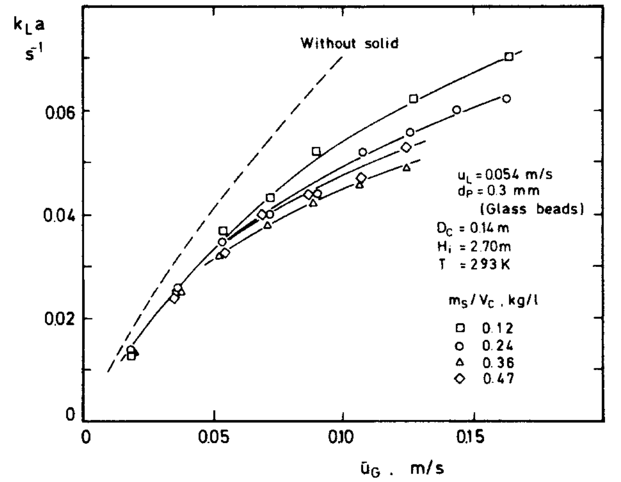

Figure 6-a.4. Influence of solid concentration on k_La decrease (Nguyen-Tien and Deckwer, 1981).

Voigt et al. (1979, 1980) and Hecht et al. (1980) have studied the mass transfer in a multistage bubble column for electrolyte solutions, highly viscous Newtonian and non-Newtonian (carboxy methyl cellulose and polyacrylate) solutions. They have recommended use of multistage columns for a coalescence promoting medium, though for non-Newtonian solutions better mass transfer can be achieved in single stage bubble columns (Voigt et al., 1980). Fan et al. (1975) and Wang and Fan (1978) studied the oxygen transfer in a bubble column packed with Koch motionless mixers. They reported a significant increase (as high as two fold) in k_La and dependence of k_La on the liquid flow rate. Hsu et al. (1975) reported higher k_La values with Koch mixers than the ones with the sieve trays in bubble columns. Orazen and Erickson (1979) reported significantly larger k_La in the two-stage airlift tower for a gas velocity between 0.20–0.45 m/s. No difference in k_La was apparent at superficial gas velocity from 0.12–0.20 m/s. Lin et al. (1976) have shown that in the tower cycling fermenters for volatile substrates, higher mass transfer rates can be achieved. Fujie et al. (1980) observed higher volumetric oxygen transfer coefficients in the downflow bubble column compared to the ones in the conventional upflow systems. No correlation for the volumetric mass transfer coefficient has been reported in anyone of the above modified bubble columns.

For prediction of k_La, equations of type (6-a.2) can be recommended with $n = 0.8$, but the constant b depends sensitively on the type of sparger and liquid media. Some values of b are given in Table 4-a.1. The majority of all the reported data indicate that k_La is not affected by the liquid flow rate if the evaluation of raw data is done by means of realistic models. Akita and Yoshida's correlation (1974) can be recommended as a conservative estimate applicable for the less effective spargers. Such spargers are also used in the multistage bubble columns for redistribution of gas and in such cases Akita and Yoshida's correlation applies. As k_La is not affected by u_L, the given correlation should also apply to bubble columns with liquid circulation (loop reactors). Its applicability has been checked for large diameter columns. For highly viscous non-Newtonian media the correlation of Deckwer et al. (1981d) is applicable if slug flow prevails. This correlation has been checked with real fermentation broths of pronounced non-Newtonian behavior. No correlation is available to estimate the effect of the presence of solids on k_La.

Liquid Side Mass Transfer Coefficient k_L

For mass transfer processes accompanied by slow chemical reaction, a knowledge of k_La is sufficient. For fast and instantaneous chemical reaction within the liquid film, a knowledge of k_L is required to calculate the enhancement factors. Owing to the uncertainties in interfacial area determinations (Weisweiler and Rosch, 1978; Schumpe and Deckwer, 1980b), the estimation of k_L from a knowledge of k_La and measured a values may involve large errors. k_L data can be determined from measurements with single bubbles. Recent work (Hallensleben, 1980) shows that results for single bubbles can be used for bubble swarms provided the flow regime is bubbly ($u_G \leq 0.06$ m/s) and the mass transfer of pure gases with low solubility is concerned. If gases of high solubility are taken into consideration, high mass transfer rates occur and the situation is much more complex (Deckwer et al., 1978; Hallensleben, 1980). Calderbank and Moo-Young (1961); Hughmark (1967); Reuβ (1970); Akita and Yoshida (1974), Gestrich et al., (1976); and Schügerl et al. (1977) have proposed correlations for k_L or Sherwood number.

Some of the widely used correlations for k_L or Sherwood number are listed in Table 4-b.1. The empirical equations developed by Calderbank and Moo-Young (1961) and by Hughmark (1967) are widely used and their application is also recommended. In the presence of solids an increase in k_L is expected (Kato et al., 1973).

Gas Side Mass Transfer Coefficient k_G

Gas side resistance to mass transfer becomes important during the absorption of highly soluble lean gas accompanied by instantaneous and irreversible chemical reaction. ven den Berg and Hoormstra (1977) have shown that the gas side resistances are present in the absorption of chlorine in benzene, even for gas mixtures with about 50% volume of chlorine. Absorption of lean SO_2 or Cl_2 or I_2 into NaOH solution or absorption of lean NH_3 or triethylamine into sulfuric acid are other examples where the gas side resistances are important. Braulick et al. (1965), Mashelkar and Sharma (1970), Kato et al. (1973), and Botton et al. (1980) have reported k_Ga data for bubble columns. Botton et al. (1980) found that k_Ga varies as $u_G{}^{0.75}$ up to a gas velocity of 0.1 m/s. Mashelkar and Sharma (1970) have covered a gas velocity range of 0.1 m/s to 0.34 m/s and they found an increase in k_Ga with u_G. Mashelkar and Sharma (1970), Botton et al. (1980) found k_Ga to be independent of liquid velocity and it decreases with an increase in the emulsion height. Kato et al. (1973) found that at low gas velocities for 0.1 m diameter column k_Ga decreases with an increase in average particle diameter, average concentration of suspended solids, and hole diameter of the gas distributor. With an increase in gas velocity, k_Ga becomes gradually independent of all the three parameters. For column diameter greater than 0.2 m, the effect of mean diameter of the particle diminishes. No correlation for k_Ga in bubble column reactor has been reported and at this stage no recommendation can be given.

TABLE 4b-1. REPORTED STUDIES ON k_L

	System	Range of Parameters	Correlation	Reference
1.	(A) for large bubbles aqueous glycol solutions-CO_2 Water-CO_2 Water-O_2 Brine-O_2 Polyacrylamide solution-CO_2	ρ_L (kg/m^3): 1,000–1,178 μ_L (Pa·s) 0.0006–0.0897 $\rho_L - \rho_G$(kg/m^3): 1000–1178 $d_b > 2.5$ mm	$k_L = 0.0042 \left(\frac{(\rho_L - \rho_G)\mu_L g}{\rho_L^2}\right)^{1/3} * \left(\frac{\mu_L}{\rho_L D_i}\right)^{-1/2}$	Calderbank and Moo-Young (1961)
	(B) for small bubbles aqueous glycol solutions-CO_2 Brine-O_2 Wax-H_2 Aqueous ethanol solution —C_6H_6	ρ_L (kg/m^3): 698–1160 $\rho_L - \rho_G$ (kg/m^3): 174–1160 μ_L (Pa·s): 0.00084–0.001 $d_b < 2.5$ mm	$k_L = 0.0031 \left(\frac{(\rho_L - \rho_G)\mu_L g}{\rho_L^2}\right)^{1/3} * \left(\frac{\mu_L}{\rho_L D_i}\right)^{-1/3}$	
2.	Air-Water Air-Na_2CO_3 soln. Air-Varsol Air-Glycerol Solutions Air-$ZnCl_2$ Solution	D_C (m): 0.025–1.1 ρ_L (kg/m^3): 776–1696 σ (N/m): 0.025–0.076 μ_L (Pa·s): 0.0009–0.152	$\frac{k_L d_b}{D_i} = 2 + 0.0187 \left[\mathrm{Sc}^{0.339}\mathrm{Re}^{0.484} * \left(\frac{d_{vs} g^{1/3}}{D_i^{2/3}}\right)^{0.072}\right]^{1.61}$ $\mathrm{Sc} = \frac{\mu_L}{\rho_L D_i}$ $\mathrm{Re} = \frac{d_{vs} u_G \rho_L}{\mu_L}$	Hughmark (1967)
3.	Water-Air Glycol-Air Methanol-Air Aqueous Solutions of Glycol-Air Aqueous Solutions of Methanol-Air Water-O_2 Water-He Water-CO_2	$u_G \left(\frac{m}{s}\right)$: 0.003–0.4 $u_L \left(\frac{m}{s}\right)$: 0.0–0.44 D_C(m): 0.152–0.6 H_i(m): 0.126–0.35 ρ_L(kg/m^3): 800–1600 μ_L(Pa·s): 0.00058–0.021 σ_L(N/m): 0.022–0.0742	$k_L = 0.5 \frac{D_i}{d_{vs}} \left(\frac{\nu_L}{D_i}\right)^{1/2} \left(\frac{g d_{vs}^3}{\nu_L^2}\right)^{1/4} \left(\frac{g d_{vs}^2 \rho_L}{\sigma}\right)^{3/8}$	Akita and Yoshida (1974)
4.	Aqueous alcohol solutions-Air Electrolytes-Air	$u_G \left(\frac{m}{s}\right)$: < 0.08 ρ_L (kg/m^3): 996–1,006 σ_L (N/m): 0.053–0.073 μ_L(Pa·s): 0.00089–0.001	$k_L = \frac{0.15 D_i}{d_{vs}} \left(\frac{\nu_L}{D_i}\right)^{1/2} \left(\frac{d_{vs} u_G \rho_L}{\mu_L}\right)^{3/4}$	Schügerl et al. (1978)

Liquid-Solid Mass Transfer Coefficient

In the three phase bubble column slurry reactors, the mass transfer from bulk liquid to the solid surface can play an important role in the overall apparent reaction rate (Choudhari and Ramchandran, 1980). Liquid-solid mass transfer has been critically reviewed by Shah (1979). The theory of liquid-solid mass transfer is based on the slip velocity (boundary layer) between two phases, and in general, leads to expressions like,

$$\mathrm{Sh} = 2.0 + \alpha \mathrm{Sc}^m \mathrm{Re}^n \qquad (6\text{-d.1})$$

Frössling (1938), and Ranz and Marshall (1952) showed that for large values of Re, $\alpha = 0.6$, $m = 1/3$ and $n = 1/2$. Shah (1979) has discussed in details the recommendations of various investigators (Friedländer, 1957, 1961; Harriot, 1962; Levich, 1962; Brian and Hales, 1969) to evaluate the particle Reynolds number

$$\mathrm{Re} = \frac{u_r d_p}{\nu} \qquad (6\text{-d.2})$$

where u_r is the relative velocity between two phases. In the case of bubble columns where the solid particles are suspended, this velocity is difficult to determine.

Kolmogoroff's theory of isotropic turbulence avoids this problem by defining the length scale of the micro-scale eddies, which is defined as

$$\eta = \left(\frac{\nu^3}{e}\right)^{1/4} \qquad (6\text{-d.3})$$

and the velocity scale is defined as

$$v_1 = (\nu e)^{1/4} \qquad (6\text{-d.4})$$

where e is the local energy dissipation rate per unit mass. From the stochastic behavior of the fluid flow around the suspended particle and Kolmogoroff's theory of isotropic turbulence the following relations for the Reynolds number can be derived.

$$H_i > \eta > d_p \quad \mathrm{Re} = c^* \left(\frac{e d_p^4}{\nu^3}\right)^{1/2} \qquad (6\text{-d.5})$$

$$H_i > d_p > \eta \quad \mathrm{Re} = c^* \left(\frac{e d_p^4}{\nu^3}\right)^{1/3} \qquad (6\text{-d.6})$$

where c^* represents a dimensionless constant and H_i is a characteristic length, for instance, the suspension height. By using Kolmogoroff's approach, the energy dissipation rate in bubble columns can be calculated from the pressure drop experienced by the gas flow rate. The energy input P' is approximately given by,

$$P' = u_G \Delta p = u_G \rho_L g \epsilon_L H_i \qquad (6\text{-d.7})$$

where Δp is the pressure drop in the column. Therefore the specific energy dissipation rate can be calculated as,

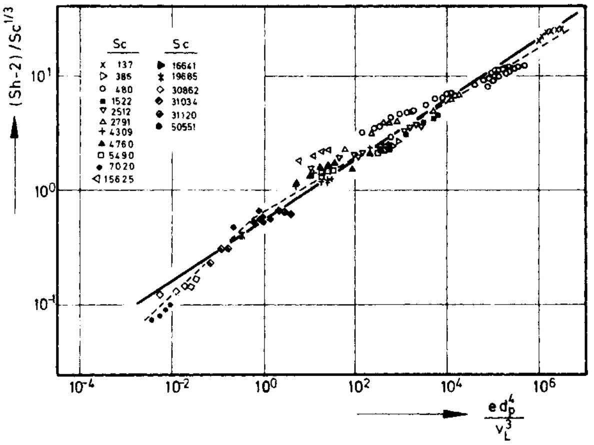

Figure 6-d.1. Correlation of liquid-solid mass transfer data in bubble column (Sänger and Deckwer, 1981).

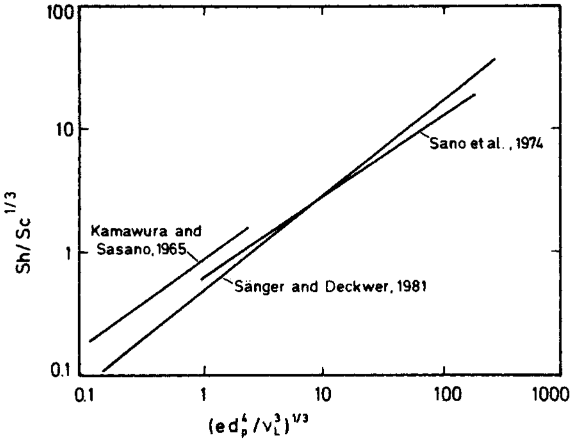

Figure 6-d.2. Correlations of liquid-solid mass transfer coefficients in bubble columns.

$$e = u_G g \qquad (6\text{-d}.8)$$

Only a few experimental studies (Kamawura and Sasano, 1965; Sano et al., 1974; Sänger and Deckwer, 1981) have been carried out in three phase vertical bubble columns. These investigations and the proposed correlations using Kolmogoroff's characteristic parameters are listed in Table 6-d.1. The data of Sano et al. (1974) covers a range of Schmidt numbers from 200 to 1400. Recently Sänger and Deckwer (1981) provided the data for Schmidt numbers ranging from 137 to 50,000. Figure 6-d.1 shows a plot of their data in Newtonian liquids. Though all the data can be better described by two regression lines as indicated by dotted lines, it is believed that one equation which covers a range of ($e\, d_p^4/\nu^3$) from 10^{-3} to 10^7 will be sufficient. The measured data deviate at low Reynolds numbers because the assumption of isotropic turbulence is not justified as discussed by Levins and Glastonbury (1972). Figure 6-d.2 illustrates that the data of three different investigators given in Table 4-d.1 are consistent. The difference in three curves is partly due to the uncertainties in the estimation of diffusivities. The figure also demonstrates the usefulness and applicability of the theory of isotropic turbulence. This is advantageous as the energy dissipation rate can be easily measured in bubble columns.

The equation by Sänger and Deckwer (1981) is recommended for the calculation of liquid-solid mass transfer since it covers a wide range of the experimental parameters.

BACKMIXING

The extent of the backmixing or the dispersion coefficient of the liquid phase in gas-liquid and gas-liquid-solid cocurrent and countercurrent flow has been the subject of many investigations. The investigations have been extensively reviewed by Shah et al. (1978b), and therefore, will not be described here. Only recent developments will be outlined.

The reported data on the liquid phase dispersion coefficient D_L indicate that D_L depends on the gas velocity and column diameter. The influence of the liquid phase properties is not clearly understood. For Newtonian liquids the effect is small and can usually be neglected. The effect of flow direction is not observed. The effect of liquid velocity, within the range of industrial interest (< 0.03 m/s) is insignificant, though at high liquid velocities, Kara et al. (1981) observed some effect of liquid velocity.

Shah et al. (1978b) recommended application of the correlations of Deckwer et al. (1974) and Joshi and Sharma (1979) which describe the literature data equally well. The various correlations described in the literature show dependencies of D_L on the diameter which range from $D_C^{1.0}$ to $D_C^{1.5}$ and on the gas velocity which varies from $u_G^{0.3}$ to $u_G^{2.0}$. Mashelkar and Ramchandran (1975) theoretically explained the dependence of the dispersion coefficient on the gas velocity in the chain bubbling and found the exponent of gas velocity to be 2.0. Recently, Rice et al. (1980) attributed this large variation to the different flow regimes. They proposed that,

- in chain bubbling, $D_L \alpha u_G^2$
- in bubbly flow, $D_L \alpha u_G$
- in churn-turbulence, $D_L \alpha u_G^{1/3}$
- in slug flow, $D_L \alpha u_G^0$

However, additional experimental evidence is required to clarify this point.

TALBE 4-d.1. REPORTED STUDIES FOR SOLID-LIQUID MASS TRANSFER COEFFICIENTS IN AERATED BUBBLE COLUMNS

System	Schmidt Number Range	Correlation Proposed	Investigator
1. Water—Benzoic Acid —$CaCO_3$ —$BaCO_3$ —PbO_2	710–1025	$Sh^* = 0.72 \left(\frac{e d_p{}^4}{3}\right)^{0.208} Sc^{1/3*}$	Kamawura and Sasano (1965)
2. Water—Benzoic Acid HCl —Napthanol —Ionic Resins	217–1410	$Sh = \Phi \left(2 + 0.4\, Sc^{1/3} \left(\frac{e d_p{}^4}{\nu^3}\right)^{1/4}\right)$	Sano et al. (1974)
3. Water Glycol Glucose—Ionic Resins Polyethylene glycol CMC	137–50000	$Sh = 2.0 + 0.545\, Sc^{1/3} \left(\frac{e d_p{}^4}{\nu^3}\right)^{0.264}$ $^* Sh = \frac{k_s d_p}{D_i}$ $Sc = \frac{\mu_L}{\rho_L D_i}$	Sänger and Deckwer (1981)

The original correlation of Joshi and Sharma (1976) recommended by Shah et al. (1978b) has been improved on the basis of a new model, i.e., the multiple circulation cell model of a bubble column (Joshi and Sharma, 1979). Along with the column diameter, the circulation velocity which is a result of dissipation of energy from gas phase, is the main factor determining the liquid phase dispersion. Joshi (1980) considered the height to diameter ratio of the circulation cell and the column to be 0.8 rather than 1.0. He proposed a correlation, which is given by

$$D_L = 0.33\,(V_C + u_L)D_C \tag{7.1}$$

where the circulation velocity is calculated from

$$V_C = 1.31\left[gD_C\left(u_G - \frac{\epsilon_G}{1-\epsilon_G}u_L - \epsilon_G u_{b\infty}\right)\right]^{1/3} \tag{7.2}$$

The dimensions are V_C, u_L, u_G, $u_{b\infty}$ in m/s, D_C in m, and D_L m^2/s. Equation 7.2 describes experimental data of various authors with a mean deviation of 10%. Field and Davidson (1980) measured the liquid phase dispersion in an industrial bubble column of 3.2 m diameter by 19 m height. Their results confirmed the applicability of the correlation of Deckwer et al. (1974) and Joshi (1980) for estimating D_L.

Riquarts (1981) made use of another theoretical approach. In analogy to the well known Pe = 2 relation for fixed beds, Riquarts assumed that this relation is also valid for bubble columns. The liquid phase dispersion coefficient is given by

$$D_L = \frac{1}{2}u_{b\infty}d_b \tag{7.3}$$

By introducing appropriate expressions for the bubble rise velocity and bubble diameter, he finally arrived at the following equation

$$\mathrm{Bo} = 14.7\left(\frac{\mathrm{Fr}^3}{\mathrm{Re}}\right)^{1/8} \tag{7.4}$$

where

$$\mathrm{Bo} = \frac{u_L D_C}{D_L} \tag{7.5}$$

$$\mathrm{Re} = \frac{u_L D_C}{\nu_L} \tag{7.6}$$

$$\mathrm{Fr} = \frac{u_L^2}{gD_C} \tag{7.7}$$

Equation 7.4 describes measured data with similar accuracy as other correlations reported in the literature.

The effect of solid particles on the liquid dispersion coefficient is not clearly understood due to the lack of enough experimental evidence. At high solid concentrations, Ying et al. (1980b) observed large discrepancies between actual and theoretical values of dispersion coefficients predicted with the help of different existing correlations. They also observed the effect of liquid velocity on dispersion coefficients. The observations were supported by Kara et al. (1981) who noted that the dispersion coefficients are lower at high liquid velocities compared to those observed in the absence of solids.

Mixing in the dispersed gas phase was measured by Diboun and Schügerl (1967), Carleton et al. (1967), Towell and Ackerman (1972), Pilhofer et al. (1978), Mangartz and Pilhofer (1980), and Field and Davidson (1980). Compared to liquid phase dispersion, this data revealed considerable scatter. Towell and Ackerman (1972) proposed the following correlation,

$$D_G = 20\,D_C^2 u_G \tag{7.8}$$

while Diboun and Schügerl (1967) proposed

$$\mathrm{Pe}_G = \frac{u_r D_C}{D_G} = 0.2 \tag{7.9}$$

Based on their recent data, Pilhofer et al. (1978) and Mangartz and Pilhofer (1980) have concluded that the bubble rise velocity in the swarm is a characteristic variable which is mainly responsible for the gas dispersion. Based on their data in different liquid media, Pilhofer et al. (1978) proposed,

$$D_G \alpha D_C^{1.5}\,(u_G/\epsilon_G)^{3.56} \tag{7.10}$$

Based on the available literature data, Mangartz and Pilhofer (1980) presented the following correlation,

$$D_G = 50D_C^{1.5}(u_G/\epsilon_G)^3 \tag{7.11}$$

Field and Davidson (1980) who conducted measurements on gas phase dispersion in a large diameter (thirty-two times that used by Pilhofer et al. (1978)) column modified slightly the original equation of Pilhofer et al. (1978) and proposed the empirical correlation

$$D_G = 56.4D_C^{1.33}(u_G/\epsilon_G)^{3.56} \tag{7.12}$$

In Eqs. 7.8, 7.9 and 7.12, D_G is in m^2/s, D_C in m, and u_G in m/s. Though the predictions of Eqs. 7.11 and 7.12 may differ up to 50%, both correlations are recommended due to lack of sufficient literature data.

By measuring the concentration profiles under cocurrent and batch conditions, Kato et al. (1972 a,b) proposed a correlation for solid dispersion as follows,

$$\mathrm{Pe}_S = \frac{u_G D_C}{D_S} = \frac{13(u_G/\sqrt{gD_C})}{1 + 8(u_G/\sqrt{gD_c})^{0.85}} \tag{7.13}$$

This equation is similar to the correlation of Kato and Nishiwaki (1972) for the liquid phase dispersion coefficient. Indeed, D_L and D_S differ only slightly. As a guideline it can be assumed that $D_L \cong D_S$. At low Froude number and large particle diameters Kato et al. (1972a,b) observed deviations from Eq. 7.13 which could be eliminated by introducing a particle Reynolds number

$$\mathrm{Re}_S = \frac{d_p u_t}{\nu_L} \tag{7.14}$$

and modifying Eq. 7.13 to

$$\mathrm{Pe}_S = \left[1 + 0.009\,\mathrm{Re}_S\left(\frac{u_G}{\sqrt{gD_C}}\right)^{-0.8}\right]\frac{13(u_G/\sqrt{gD_C})}{1 + 8(u_G/\sqrt{gD_C})^{0.85}} \tag{7.15}$$

The above correlation is based on extensive measurements in three phase bubble columns of different diameters. Therefore, use of this correlation is recommended.

Studies on the backmixing characteristics in the presence of internals are reviewed by Shah et al. (1978b). They recommend the equation by Sekizawa and Kubota (1974) and Kubota and Sekizawa (1976) for the estimation of the liquid phase dispersion coefficients in bubble columns partitioned by perforated plates, and the graphical correlation by Bischoff and Phillips (1966) in bubble columns partitioned by orifice plates. For air-water system, Blass and Cornelius (1977) proposed an empirical equation in the presence of perforated plates, for multistage bubble columns. The equation can be used to calculate the effective number of plates to achieve the desired value of Peclet number. It is as follows,

$$\frac{N_{\mathrm{eff}}}{N'} = 1.069\left(\frac{u_G{}^3_{\phi}}{u_L{}^2}\right)^{-0.19} \tag{7.16}$$

where ϕ is the fractional open area. For packed bubble columns, Magnussen and Schumacher (1978) observed that an inhibition of axial mixing is possible only by using unusually small size packing.

HEAT TRANSFER

Heat transfer from the reactor wall and inserted rods and coils to the vertical flowing gas in liquid dispersion has been the subject of many investigations and suitable correlations have been reported (Kölbel et al., 1958, 1960; Müller, 1958; Martins, 1959; Kast, 1963; Shaykhutdinov et al., 1971; Burkel, 1972; Mersmann, 1975; Hart, 1976; Steiff and Weinspach, 1978; Deckwer, 1980; Deckwer et al., 1980a; Joshi et al., 1980; Kato et al., 1981). A summary of the

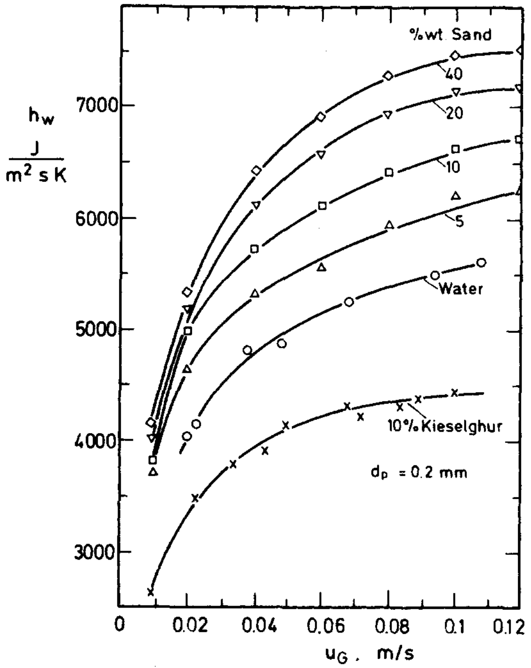

Figure 8.1. Effect of particle concentrations on wall heat transfer coefficient in three phase bubble column (Kölbel et al., 1960).

published data collected on bubble columns and stirred vessels is presented by Steiff and Weinspach (1978).

The heat transfer coefficients in bubble columns are many times larger than that for single phase flow. Kast (1963) was the first who noticed that the usual concept of heat transfer through the boundary layer does not apply to bubble agitated systems, and he related the heat transfer coefficient to the fluid motion caused by the rising bubbles. Kast (1963) pointed out that the radial component of the liquid velocity; which is induced by the rising bubble, is responsible for the high heat transfer coefficients in bubble columns. Deckwer (1980) presented a theoretical heat transfer model by combining the surface renewal model of mass transfer with Kolmogoroff's theory of isotropic turbulence. The final result in dimensionless form can be expressed as,

$$\mathrm{St} = 0.1\,(\mathrm{Re}\,\mathrm{Fr}'_G\,\mathrm{Pr}^2)^{-1/4} \tag{8.1}$$

where

$$\mathrm{St} = \frac{h_w}{\rho_L C_p u_G},\quad \mathrm{Re} = \frac{u_G d_{vs} \rho_L}{\mu_L}$$

$$\mathrm{Fr}'_G = \frac{u_G^2}{g d_{vs}},\quad \mathrm{Pr} = \frac{C_p \mu_L}{K}$$

This equation describes the data of various authors (Perner, 1960; Müller, 1962; Kast, 1963; Burkel, 1972) with striking agreement for the Prandtl number range of 6 to 985.

Heat transfer in bubble columns with suspended solids was studied by Kölbel and co-workers (1958, 1960). These authors used kieselghur in water, spindle oil and machine oil and sand particles of 40, 75, 110 and 200 μm diameter suspended in water. Some results are presented in Figures 8.1 and 8.2. Due to swelling, suspensions of kieselghur were highly viscous, which reduced heat transfer coefficients considerably. However, suspensions of sand increased the heat transfer. The heat transfer coefficients increased with increasing solid concentration and particle size. Deckwer et al. (1980a) reported the measurements of heat transfer in slurries of Al_2O_3 particles in molten paraffin wax in a 0.1 m diameter column for the purpose of simulation of heat transfer conditions encountered in Fischer-Tropsch slurry process. Figure 8.3 shows that the heat transfer data of Deckwer et al. and of Kölbel and co-workers including the data for sand particles up to 110 μm diameter can be well represented by Eq. 8.1, provided the appropriate physico-chemical properties of the slurry are used. Therefore, this correlation can be recommended for h_w calculations in gas-liquid systems including particles up to about 100 μm in size.

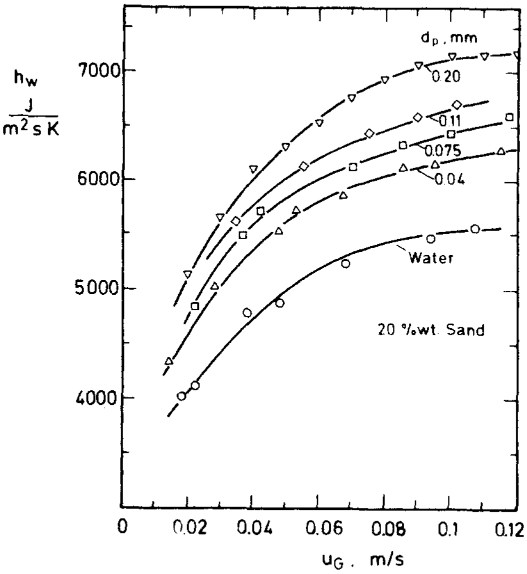

Figure 8.2. Effect of particle size on h_w (Kölbel et al., 1960).

Nishikawa et al. (1977) measured the heat transfer coefficient from reactor wall (h_w) and from immersed coils (h_c) for various Newtonian and non-Newtonian liquids. In the Newtonian liquids both the h_w and h_c values are the same. For non-Newtonian liquids, as the viscosity of the solution depends on the shear rate, the authors used heat transfer measurements to calculate the apparent viscosities and shear rates of the aerated non-Newtonian liquids. They found that at gas velocities below 0.04 m/s the value of h_c is much greater than h_w. It is therefore advantageous to use coils for the thermal control of bubble column reactors. This difference is attributed to the different shear rates at low gas velocities indicating that only the central part of column, where the cooling coil was installed, was aerated more efficiently. If $u_G > 0.04$ m/s, turbulent energy is also transported to the wall, and shear rates are equal over the cross-sectional area.

For three phase fluidized bubble columns, many investigators (Ostergaard, 1964; Viswanathan et al., 1965; Armstrong et al., 1976) have reported wall bed heat transfer coefficient values. The results of these investigators are critically reviewed by Shah (1979).

Recently Joshi et al. (1980) proposed a new correlation based on the circulation cell model (Joshi, 1980). They reported that in some gas-liquid-solid systems, all the input energy due to the gas flow is dissipated at the gas-liquid and solid-liquid interface, rather than in the liquid motion. By using the method of average recirculation velocity (Joshi, 1980), they proposed the following equation to calculate the wall bed heat transfer coefficient.

$$\frac{h_w D_c}{K} = 0.48\left[\frac{D_C{}^{1.33} g^{0.33} (u_G - \epsilon_G u_{b\infty})_\rho^{1/3}}{\mu_L}\right]\left(\frac{C_p \mu_L}{K}\right)^{1/3} \times \left(\frac{\mu_L}{\mu_w}\right)^{0.14} \tag{8.2}$$

This equation is based on the analogy between mechanically agitated contactors and bubble columns. Figure 8.4 shows a comparison between the experimental values of h_w and those predicted by above equation.

It can be seen from Figures 8.3 and 8.4 that both the correlations represent the literature data fairly well. It can therefore be concluded that the apparent discrepancy between the deterministic (Joshi, 1980) and the stochastic (Deckwer, 1980) approach is immaterial and both physical concepts are equally capable of representing transfer phenomena in the liquid phase of bubble columns, and can be regarded as limiting cases of a more general model.

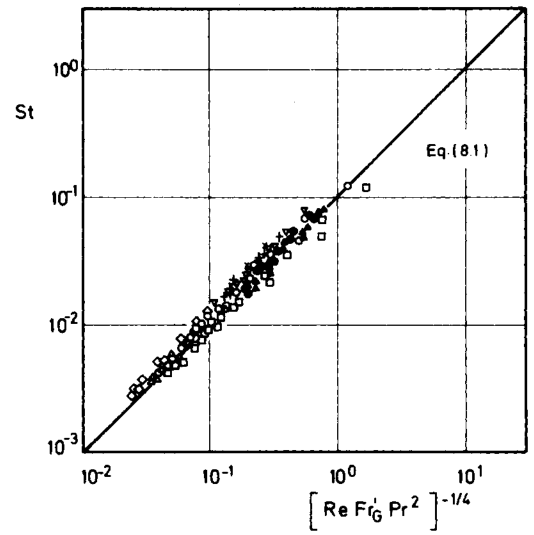

Figure 8.3. Correlation of heat transfer coefficient in slurry bubble columns by Eq. 8.1 (Deckwer et al., 1980a).

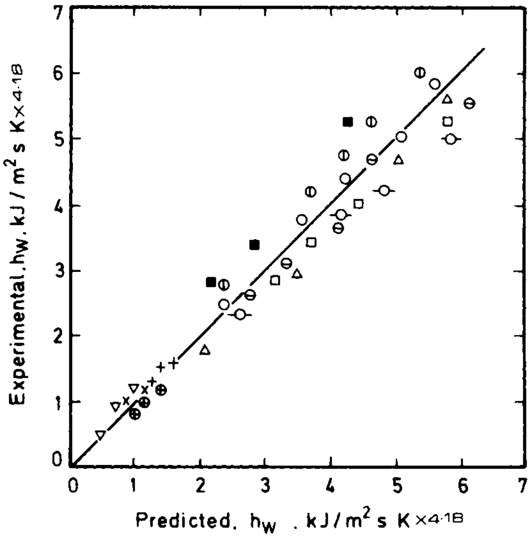

Figure 8.4. Comparison of experimental data and Eq. 8.2 (after Joshi et al., 1980).

RECOMMENDATIONS FOR FUTURE WORK

In bubble column reactors, there are many areas in which the hydrodynamic, mixing and the transport characteristics are not clearly understood. Future research should include the following studies.

1. Only approximate flow regime charts are available for liquids like water, electrolyte solutions etc. For the design and scaleup of bubble column reactors, effects of fluid properties and the nature of the gas distributor on the flow regimes need to be examined. In the study of the effect of fluid properties, the following liquids should be emphasized,
   A. dilute alcohol solutions
   B. organic liquid media
   C. highly viscous slurries
   D. non-Newtonian liquids
2. It has been pointed out in the literature that modified bubble columns are sometimes advantageous over single staged bubble columns. Unfortunately, insufficient data are available for novel reactors for any specific design recommendations. Following novel reactors should be studied in more details:
   A. loop reactor
   B. jet reactor
   C. staged bubble column
   D. packed bubble column
   E. countercurrent bubble column
   F. downflow bubble column
3. Due to the complex flow characteristics, the energy dissipation in the bubble column has been the subject of many investigations. Various models have been proposed to explain this dissipation; the most widely used being the Kolmogoroff's isotropic turbulence model. Although most of the time this theoretical approach is camouflaged by too much empiricism, the validity of this isotropic turbulence model should be checked in more detail, especially at low gas velocities.
4. Gas holdup and interfacial area often depend on fluid properties other than ρ, σ or μ, even for Newtonian fluids. More data are needed for non-aqueous organic media like alcohols, ketones, esters and hydrocarbons and gases such as hydrogen, chlorine, etc., in large diameter bubble columns.
5. Many hydroprocessing operations are carried out in high pressure, high temperature bubble columns. Some data on hydrodynamics, mixing and transport characteristics under these conditions with real systems should be obtained.
6. Many times liquid phase is observed to foam in the presence of gas resulting in excessively high values of gas holdup. The stability of foam under different sets of parametric conditions is not yet clearly understood, therefore, it should be evaluated in more detail.

NOTATION

a = specific gas-liquid interfacial area, m^{-1}
b = constant in Eq. 6-a.2
Bd = Bond number, defined in Eq. 3.3
Bo = Bodestein number, defined in Eq. 7.6
C_p = specific heat of liquid phase, J/kg·K
d_b = individual bubble diameter, m
d_p = particle diameter, m
d_{vs} = Sauter mean bubble diameter, m
D = axial dispersion coefficient, m^2/s
D_C = column diameter, m
D_l = molecular diffusivity of solute in liquid phase, m^2/s
e = local energy dissipation rate per unit mass, J/kg·s
e' = energy dissipation rate, J/s
Eo = Eötvos number, defined in Eq. 3.2
f_1 = function defined in Eq. 2.3
f_2 = function defined in Eq. 2.4
Fr = Froude number, defined in Eq. 7.8
Fr'_G = Froude number for gas phase, defined in Eq. 8.1
g = gravitational acceleration, m^2/s
g_c = gravitational constant
G = gas phase inlet
h_c = coil bed heat transfer coefficient, $J/m^2{\cdot}s{\cdot}K$
h_w = wall bed heat transfer coefficient, $J/m^2{\cdot}s{\cdot}K$
H = Henry's constant, $Pa{\cdot}m^3/kg$ mol
H_t = dispersion height, m
j_{GL} = drift flux velocity, m/s
k = mass transfer coefficient, m/s
k_m = rate constant
k^*_m = absorption reaction parameter, defined in Eq. 5.6
K = thermal conductivity of liquid phase J/m·s·K
K' = dimensional constant in Eq. 5.8
L = liquid phase inlet
m = order of reaction
m_s = mass of the solids in the reactor as in Figures 6-a.3 and 6-a.4, kg
M = Morton number, defined in Eq. 3.7
N = number of bubbles having diameter d_b
N' = actual number of plates in multistage bubble column
N_{eff} = effective number of plates in multistage bubble column
P = pressure, Pa
P' = energy input, J

Pe_G = gas phase Peclet number, defined in Eq. 7.10
Pe_S = solid phase Peclet number, defined in Eq. 7.14
Pr = Prandtl number, defined in Eq. 8.1
R = gas constant, Pa m^3/kg mol·K
Re = Reynolds number, defined in Eqs. 3.7, 7.7; Table 4-b.1
Re_p = particle Reynolds number, defined in Eq. 7.15
Sc = Schmidt number, defined in Table 4-b.1, 4-d.1
Sh = Sherwood number, defined in Table 4-d.1
St = Stanton number, defined in Eq. 8.1
St_m = Stanton number, defined in Eq. 5.5
T = temperature, K
u = superficial fluid phase velocity, m/s
$u_{b\infty}$ = terminal single bubble rise velocity, m/s
u_G^* = interstitial gas phase velocity, defined in Eq. 3.5, m/s
u_r = relative velocity between two phases, m/s
u_t = terminal velocity of a solid particle, m/s
U_s = slip velocity, m/s
v_1 = velocity scale of microscale eddies, m/s
V_C = circulation velocity, defined as Eq. 7.3, m/s
V_D = dispersion volume, m^3
V_R = reactor volume, m^3
We = Weber number, defined in Eq. 3.4
X = conversion of gas phase

Greek Letters

$\dot{\gamma}$ = shear rate, s^{-1}
ΔP = pressure drop, Pa
η = length scale of microscale eddies, m
ϕ = fractional open area
Φ = Carmann surface factor
ρ = phase density, kg/m^3
σ = interfacial tension, N/m
ν = kinematic viscosity, m^2/s
ϵ = phase holdup
τ = gas phase residence time, s
μ = dynamic viscosity, Pa·s
μ_{eff} = effective viscosity calculated with the help of power law model, Pa·s
μ_w = viscosity of water, Pa·s

Subscripts

G = gas phase
L = liquid phase
S = solid phase
SL = slurry phase
O = inlet
1 = outlet

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Manuscript received February 4, 1981; revision received January 15, and accepted January 28, 1982.