MASS TRANSFER IN FLUIDIZED BEDS

by

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ABSTRACT

Three internal mass transfer problems and one dealing with slow bubble properties are analyzed.

1. A literature survey was made of the different bed models applied to bubble to emulsion mass transfer: of the many fast bubble to emulsion models, a convective model, by Davidson and Harrison, and two diffusion-type models, by Kunii and Levenspiel and Chiba and Kobayashi are compared. Of twenty papers with experimental data, only four are adequate for analysis, and only one of these presented sufficient data for direct calculation of the mass transfer coefficient. All mass transfer coefficient data for minimally fluidized beds is plotted together to compare the three models.

The Davidson and Harrison model is better for large particles (>500μ) and for overlapping clouds. The diffusion-type models are preferable for smaller particles. Other mechanisms are suggested for mass transfer.

2. It is shown analytically that modest size slow bubbles are good mixing points. A model is constructed which assumes all gas is radially dispersed by stationary slow bubbles. An effective radial dispersion coefficient is calculated and compared to those found experimentally in fast bubbling beds, showing that generally the former are larger. No experimental work has been found for slow bubbles.

3. A study on the proper values of Sh and Nu for individual particles in fixed and fluidized beds is presented. Most explanations of the phenomena rely on models invalid at Reₚ of unity and below. The actual limiting value of Sh and Nu is shown to be equal or greater than unity. A model of a typical experimental system was established. The observed fall-off in Sh and Nu derived from experimental data is found to be caused by improper accounting for, or omission of, phenomena important at low Reₚ.

4. Geometric properties of the recirculation region and the throughput coefficient for slow bubbles are evaluated for various bubble arrangements. These are combined for the case of a swarm of three dimensional bubbles to form the effective throughput coefficient M. It is found that M = 3 for a large range of conditions.

Thesis Advisor: Leon R. Glicksman
Title: Mechanical Engineering Lecturer
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CHAPTER I

1 Introduction

"A fluidized bed is a bed of particles on a horizontal gauze or distributor through which there is an upward flow of fluid sufficient to support the weight of the particles. The minimum superficial velocity to support the weight of the bed is known as the incipient fluidizing velocity, $u_{mf}$. Most gas-fluidized systems have an agitated appearance when the superficial velocity $u_o$ is greater than $u_{mf}$; this is aggregative fluidization."

(From Davidson et al [1])

Fluidization has been used for industrial purposes for over half a century. It was initially used for the gasification of powdered coal by Winkler. Since then fluidized beds have found many applications in industry such as for synthesis reactions, cracking and reforming of hydrocarbons, carbonization, gasification, calcining, clinkering and gas-solid reactions and as heat exchangers. The advantage of fluidized beds lies principally in the rapid mixing of solids and gases and the large surface contacting, leading to nearly isothermal conditions throughout the bed which make its operation simply and reliably controllable, high heat transfer rates to immersed surfaces, and a very effective means of enhancing chemical reactions.

However, the path to commercial success with fluidizing processes has proved to be unusually painful and complex, with many stages of scale up and more than its expected share of embarrassing failures. These dif-
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ficulties are due to the lack of satisfactory answers to the many questions on which design decisions are based, and this in turn stems from a lack of reliable predictive knowledge of what goes on in a bed.

Study of fluidized bed fundamentals has increased its momentum in recent years because of its advantages in coal combustion. The USA, a consumer of a third of the planet's energy and possessing one sixth of its reserves [2] has most of its energy reserves in the form of coal, part of which has a high sulphur content. Fluidized beds consisting of up to 97% limestone or dolomite and the rest coal may be an inexpensive and effective method of burning coal and reducing SO\textsubscript{x} and NO\textsubscript{x} emissions without the use of such equipment as scrubbers.

As indicated above, there are many questions which have up to date had unsatisfactory answers. Some of these lie in the mass transfer area.

For design purposes, the knowledge of the distribution of the different gaseous species within a reactor is fundamental. Whether a fluidized bed performs as a continuous flow stirred-tank reactor (CSTR) or as a plug flow tabular tank reactor (PFTR) or to which degree may it be similar to either of these extreme cases is a determining factor of its size. To determine this behavior, it is necessary to address such problems as the possible by-passing of reacting gases of the bed by traveling in the bubbles, the effect of bubbles in aiding in radial mixing of the gaseous species and the rate at which a species may leave a particle's surface. These are all mass transfer problems.
Most of the work presented in this thesis deals with fluidized beds in the aggregative state. The two-phase theory is assumed [1], which has as its major assumption that all of the gas flow in excess of that required to fluidize the particles travel through the bed as voids relatively free of particles called bubbles.

These gas bubbles are not isolated from the denser gas-particle or particulate phase. This creates a crossflow of different gas species between the bubbles and the particulate phase. Frequently high contact between the gas and the particles is required, for instance in the combustion of coal. It is quite probable that the gas within the bubbles will be rich in the gas of interest, e.g. oxygen, needed for reaction with the particles, e.g. coal, at the exit of the bed. This by-passing phenomenon should be avoided.

The crossflow problem in a fluidized bed is only important when the velocity of the bubble is faster than that of the gas in the particulate phase; this is a fast bubble. Then the gas which leaves the bubble's roof is drawn down around it and reenters it from the bottom [3]. In this situation the bubble gas is essentially an isolated system, which mixes with the rest of gas in the particulate phase by dispersion. The study of mass transfer from fast bubble to emulsion is the subject of Chapters II through IV.

First the models to be used in the analysis of experimental data and some of those found in the literature are presented and critically discussed. These models consist of a fluid mechanical model of
the bed and the mass transfer processes between bubble and particulate phase gas. Then a collection of experimental papers is scrutinized for the correlation of data and comparison to three of the most common theoretical mass exchange models. All the data found for mass transfer for minimally fluidized beds was placed on a single graph for comparison. In the last chapter an explanation is attempted for the observed phenomena. Different mechanisms are considered, such as dispersion, wakes behind particles and interactions and perturbations between neighboring bubbles. A model covering different situations and accounting some of the mechanisms suggested is proposed. Finally, recommendations for future work is made.

The size of particles used in a fluidized bed combustor will be large (>500μ) compared to that of the vast majority of the experimental work done up to now. It is expected that for the fluidization of large particle systems, the bubbles in the bed will be slow over a significant superficial velocity range [4]. Slow bubbles are those that rise at a lower velocity than the gas in the particulate phase. In this case a fraction of the gas entering the bubble flows right through it while the rest leaves the bubble to reenter it later on, as in the fast bubble. It is of interest to find out how well these bubbles act to disperse the gases within the bed, assuming that they act as perfect mixers. Consequently a simple model of stationary slow bubbles was created to estimate the radial dispersion produced by these bubbles. As no experimental work has been found in this subject, the results were
compared to radial dispersion in fast bubbling beds.

A third topic considered was mass transfer from particles to the surrounding fluid. Frequently chemcal reactions in a fluidized bed are meant to occur between a gaseous species and another which is originally in solid form. If the chemical kinetics of the process are rapid enough, it is "diffusion dominated", i.e. the reaction depends mainly on the rate at which the species in the solid vaporizes and diffuses through the film of products around the solid to the gaseous species. In fluidized beds where absorption or the reaction occurs at the level of the pores of the particles is important, the rate of diffusion of the gaseous species to the particle must be considered. Both of these effects are found in fluidized bed combustors, where coal particles are burnt and $SO_x$ reacts at the pore level with calcium oxide in the limestone or dolomite particles. Experiments in both fluidized and packed beds have been made in the last thirty years. For particle Reynolds numbers greater than 50, the trend of the Sherwood and Nusselt numbers are more or less well established. However, at lower particle Reynolds numbers experimental results have been highly controversial. Early experiments in packed beds indicated that the Sherwood number approached zero as the Reynolds number became very small. Moreover, different investigators' results varied by more than an order of magnitude. Recently some experimenters have achieved higher values of the Sherwood number. Theoreticians have attempted to justify the observed phenomenon, some by suggesting that the Sherwood number should in fact fall off to
zero and others by creating models of a bed which would describe flow patterns which would lower the Sherwood number. The latter apply unrealistic parameters or concepts and it is proved that the former are wrong, as it is shown that the actual Sherwood and Nusselt number must be larger or equal to unity.

It has been suggested that the observed fall-off in Sherwood and Nusselt numbers derived from experimental data is partly caused by improper accounting for, or omission of, phenomena important at low Reynolds numbers such as flow channeling and axial diffusion. The particulate phase in a fluidized bed has similar characteristics to that of a packed bed, so the effects observed in the latter should also be found in the former. Furthermore, inhomogeneities due to the bubbles containing gas at lower concentrations (if the transfer is from the particles to the gas) and increased axial mixing in the particulate phase due to the motion enhanced by the passing bubbles should result in even lower experimental Sherwood numbers. In fact, this is what has been experimentally observed [5]. The same may be said of the Nusselt number.

The last problem studied deals with the through-flow and recirculation properties of isolated and swarms of slow bubbles. As in most fluid mechanical problems the equation of mass conservation is fundamental in the modeling of a fluidized bed. Of particular importance is the flow related to slow bubbles. As already explained, a fraction of the gas enters the bubble and later exits into the emulsion without returning to the void. Another fraction leaves the bubble to
reenter it. As the velocity of the bubble increases, the former decreases and the latter increases. From the knowledge of these values for the simple isolated two and three dimensional bubbles and a two dimensional horizontal array of bubbles, it is possible to estimate these properties for the more realistic three dimensional swarm of bubbles. The latter are finally combined into a single effective through-flow coefficient which greatly simplify the overall mass conservation equation for a two phase fluidized bed fluid mechanical model.
REFERENCES


CHAPTER II

STUDY OF THE GAS MASS TRANSFER FROM FAST BUBBLE TO EMULSION - I

The models

II-1. Introduction

In fluidized beds with fast bubbles most of the gas superficial velocity in excess of that used to fluidize the particulate phase tends to travel through the bed in the form of bubbles. If the bubbles rise at a higher velocity than the gas in the particulate phase, they form isolated pockets of recirculating gas. Consequently, gas fed in at the distributor required for chemical reaction in the bed may then bypass the particulate phase.

In this chapter, the equations that were used for data reduction (species conservation, the fluid mechanical model, and the mass transfer equations) and many of the theoretical bubble to emulsion mass transfer models are presented and thoroughly discussed. The physical assumption of each are examined critically, frequently referring to observations and opinions of many researchers within the field.

II-2. ASSUMPTIONS FOR DATA REDUCTION

II-2.1 The Tracer Conservation Equation. In only a few of the experiments reviewed (Grace & Chavarie, 1972, 1975 and Pereira and Calderbank 1975) are the concentrations of the dense and disperse regions simultaneously measured. A mole balance on the tracer gas must therefore be made. The following assumptions are frequently (implicitly or explicitly) made.
1. No chemical reactions in the bed.
2. The emulsion phase concentration is in plug flow, i.e. radially well mixed.
3. The emulsion concentration is in steady state.
4. Individual bubbles are well mixed and are in plug flow.
5. The cloud and wake are well mixed and are in plug flow.
6. There is no axial dispersion within the emulsion phase.

Conditions (1) and (6) eliminate all possible transfer or consumption of the tracer species within the emulsion. The rest permit a time average to be taken across any plane perpendicular to the bed axis. The mole balance is

\[ F_b C_b + F_c C_c + F_e C_e = F_o C_o \]

where \( F_b \), \( F_c \) and \( F_e \) and \( C_b \), \( C_c \) and \( C_e \) are the flow rates and tracer concentrations of the gas in the bubble, cloud/wake and emulsion, respectively, and \( F_o C_o \) is the total number of moles of the tracer passing through the bed plane at that height. This value is constant and usually equal to the number of moles of trace injected at the bottom of the bed.

A seventh condition is sometimes assumed. The cloud/wake and bubble concentrations are equal. Then the above equation becomes

\[ (F_b + F_c) C_b + F_e C_e = F_o C_o \]  \hspace{1cm} \text{II-1} \]

It should be noted that in much of the work done with small clouds when they may be at the bubble concentration, \( F_c \) was taken as
\( (c_{mf}/3) F_b \), i.e. a wake fraction of 1/3 was assumed. This value seemed to be in agreement with the opinion of some of the authors such as Rowe and coworkers (1965, 1971).

II-2.2 Discussion. A discussion on the validity of the above assumptions follows.

1) Chemical reactions. Many authors (see Table III-1) have made experiments where reactions were involved. However, as will be discussed in (III-2), this increases the complexity of the processes in the bed to a point where results may very well be obscured. Consequently, reactions will not be considered.

2) The emulsion concentration is in plug flow. By plug flow it is meant that all the properties of the phase do not vary radially (see Levenspiel, 1962). According to Sherwood et al (1976), for packed beds the velocity profile may be considered to be in plug flow if the particle diameter is much smaller than the bed diameter. Then, for superficial velocities \( u_0 \) close to minimum fluidization, the emulsion phase should also be in plug flow if the particle diameter is much smaller than the bed. On the other hand Calderbank et al (1975) have indicated that they have found upward interstitial velocities in the middle of the bed and downward velocities near the wall. They were working with a freely bubbling bed, while a large portion of the data to be considered took place in beds where bubbles were injected individually and so the dense phase was possibly more homogeneous.

As the velocity profile is not flat it is difficult to expect
that the concentration will be in plug flow. In the majority of experiments considered bubbles were injected at the center of the bed at the height of the distributor, and consequently rise preferentially through the center. This necessarily causes radial concentration gradients as well as increases the mixing in the axial direction. Stephens et al (1967) showed that the concentration profiles flatten out with increasing height, and with decreasing particle size. The gradients will have the effect of reducing the calculated interchange coefficient. Walker (1975) recognizes that these are an important source of error. Latham and Potter (1970) and Kobayashi et al (1969) present profiles which are not flat for quite a distance from the injection point.

A mechanistic point of view may be taken. Rowe and Evans (1976) and Hiraki et al (1968) have studied the effect of radial dispersion in a bed. Their apparatus and methods of measurement are very similar to those which are to be treated here. The two differences are that the beds are usually wider and that tracer gas is not injected to form bubbles but directly into the emulsion phase. Following Bischoff and Levenspiel (1962), a dispersion model for the emulsion phase may be assumed. As shown in Appendix (II-1), plug flow conditions within the bed result when

\[
\frac{D_r}{(u_R)^2} \frac{D_r}{D_a} \frac{z^2}{R^2} \gg 1.
\]

If

\[
\frac{D_r}{(u_R)^2} \frac{D_r}{D_a} \frac{r^2}{R^2} \gg 1,
\]

most of the bed will be in plug flow. \(D_r\) and \(D_a\) are effective radial
and axial dispersion coefficients of the freely bubbling fluidized bed, \( u_e \) is a typical interstitial velocity, \( R \) and \( L \) the radius and height of the bed, assuming the injector is at the bottom.

The above condition rarely holds, considering that according to Hibi (1962) the interstitial dispersion coefficients should be similar to molecular diffusion at low Reynolds numbers. However, as the bubbles (which were disregarded in the above analysis) have comparable diameters to the bed diameter, it is quite possible that mixing is enhanced by them.

There are some models which include dispersion coefficients, such as Van Deemter's (1961) Mireur and Bischoff's (1967) and Drinkenburg and Rietema's (1972). Unfortunately the values used are as yet unreliable.

Due to lack of reliable predictions, it will be necessary to maintain this assumption. Admittedly, it is more accurate than assuming a well mixed bed, as was done by Davidson and Harrison (1963), and may be seen from the available experimental work. The writer agrees with Chavarie and Grace (1972), that more modeling is required in this region, considering that large beds will deviate from plug flow more than small beds.

3) The emulsion concentration is in steady state. This assumption appears to be quite adequate even though in many cases the passage of the bubbles is periodical. In fact, the bed is really in a quasi steady state regime. If bubbles pass periodically at a given height, the bubble concentration will only be a function of height. However, as in experiments usually the emulsion phase concentration is measured continuously, it is necessary to investigate this point.
The bubble and surrounding concentration profiles published by Pereira and Calderbank (1975) in Fig. (1) show that indeed, the concentration within the emulsion does rise after the passing of a bubble. However it is never more than 8%. To show that the bed is in steady state the convective derivative will be compared to the time derivative. That is:

\[
\frac{DC}{Dt} = \frac{\partial C}{\partial t} + u_e \frac{\partial C}{\partial h} \approx u_e \frac{\partial C}{\partial h}
\]

The characteristic time is the residence time of the emulsion gas in the bed, and the characteristic length the distance that the average emulsion gas travels between the appearance of two consecutive bubbles at a given height. The characteristic concentration is the total emulsion concentration difference between top and bottom of the bed.

\[
\frac{\partial C}{\partial t} \sim \frac{\Delta C}{L/u_e}, \quad u_e \frac{\partial C}{\partial h} \sim \frac{u_e \Delta C}{u_e/f_L}
\]

where \(f_L\) is the level bubble frequency and \(\Delta C\) the concentration difference. The condition then becomes

\[
\frac{u_e}{L/f_L} \ll 1
\]

This may also be interpreted as the ratio between the emulsion gas residence time within the bed and the time between the passing of two consecutive bubbles at a level in the bed. So if the number of bubbles going through the bed with one emulsion gas residence time is very large,
the emulsion concentration may be assumed to be in steady state. Still another interpretation is that of the ratio between emulsion and bubble velocities divided by the total number of bubbles in the bed.

Some experimentors such as Szekely (1962), Guedes de Cravalho and Harrison (1975) and Grace and Chavarie (1976) and Toei and coworkers (1968, 1969, 1970), Rowe et al (1971), Drinkenburg and Rietema (1973) have assumed a negligible bed concentration. In the case of the first two it appears that this is assumed because reactions taking place within the bed are fast. For the others supposedly the residence time of the gas in the emulsion phase was far shorter than the bubble injection period. In many of the papers this was not clarified. In fact, Walker (1975) suggests that in Toei's and coworker's (1968, 1969) case, this was a source of error.

4) Individual bubbles are well mixed and in plug flow. This assumption has been confirmed by a few investigators. Reuter et al (1970) worked in a two dimensional bed taking interferometer photographs of bubbles. He concluded that the concentration was constant throughout the bubble except at the bottom-most part where gas emerges from the wake. This is because the gas from the cloud/wake region should not be as rich in the bypassing gas as the rest of the bubble. Consequently, it would require a short region where it may become totally mixed. Pyle and Rose (1965) indicate that most probably the gas at the top of the bubble becomes turbulent due to an increase of the Reynolds number by at least one order of magnitude, and the lower part is in transition from viscous flow. Re-
portedly this has been confirmed by Rowe and Partridge (1965). This process would explain why the gas is well mixed in most of the bubble concentration. Profile measurements of three dimensional bubbles by Pereira and Calderbank (1975) taken at the vertical diameter with a probe, seem to confirm the above. Their probe may have interfered in the measurements, which are shown in Fig (II-1) which was taken from their paper. It is noted that in their case the bubble maintains the same concentration throughout.

Mireur and Bischoff (1967) justify the common assumption of bubbles being in plug flow by indicating that bubbles travel at high velocities with respect to the emulsion phase which impeded backmixing. Admittedly, if bubbles are of different sizes at a given height, their concentrations will be different. We will nonetheless assume that all bubbles are typical and in plug flow.

5) The cloud and wake are well mixed and in plug flow. This assumption may not be very realistic as the gas emerges from the upper part of the bubble and is swept around it, the tracer diffuses outward to the surrounding emulsion, becoming leaner on its way. Diffusion from the void surface feeds the cloud fluid at the same time, compensating for the lack of the convective part according to the Davidson and Harrison model (1963).

The only data found of direct measurements of the cloud are those by Pereira and Calderbank (1975). From their limited data it is evident that the cloud is not at a constant concentration, but more
Figure II-1  Tracer Concentration within the Bubble and its Surroundings (from Pereira & Calderbank (1975))

A = above the bubble (cloud and emulsion)
B = bubble
C = below the bubble (wake, cloud and emulsion)
approximately linear. Drinkenburg and Rietema (1972) have presented a rather sophisticated model which confirms this sort of behavior. The concentration profile is solved for in Appendix II-2. As a result, from the diffusive mass transfer viewpoint, there exists an important resistance which is not accounted for in the simpler models. Naturally, its importance is reduced if the cloud is very thin which is when, strictly speaking, the Kunii and Levenspiel (1969) model was meant to apply. However, when the ratio of bubble to emulsion velocities (α) is less than 5, the presence of the cloud should be important, not being possible to identify it with the two surface resistances.

This aspect might be of special importance when considering chemical reactions, as the concentration profiles within the cloud may be even steeper due to the depletion of the species in the cloud itself, as Drinkenburg and Rietema (1973) indicate. But, of course, this is more accurate than assuming that the cloud and either the emulsion or the bubble are at the same concentration. Whether the resistances thought to dominate this region are those that have been calculated remains open to question.

For the wake, essentially the same applies. From the concentration profiles of Pereira and Calderbank (1975), it may be concluded that there is a region of intermediate concentration between bubble and emulsion phase. Its thickness is one to two times the thickness of the region above the bubble, the latter supposedly representing the cloud. The lower part, although the evidence is hardly conclusive, may be assumed
to be the wake. On the average, the concentration in the lower region is lower than at the top. As the cloud is assumed to be at constant concentration and also present at the bottom, the wake should be modeled at a lower concentration. However, due to the lack of information, it will be assumed to be at the cloud's concentration.

6) There is no axial dispersion in the emulsion phase. Some models, already mentioned before, have included an axial dispersion term for the emulsion phase. If non-absorbent particles are used, then the importance of this term should be reduced because mass transfer can then only occur by molecular diffusion and bulk fluid motion. Nevertheless, there is some mixing which may be accounted for by a term of this type, but because of lack of knowledge it is necessary to disregard it.

If highly absorbent particles are present, it may be necessary to account for gas appearing in different parts of the bed by particle motions, and hence increasing the mixing by a term like this as suggested by Potter (1971), Van Deemter (1961) and Yates (1975). Also gas-rich or poor in the tracer may envelop a particle at low Reynolds numbers, as suggested by Happel's "free surface model" (see Pfeffer and Happel (1964)).

Fortunately in the cases studied here, only non-reactive non-absorbent particles were used, which helps justify neglecting the term. However, for future modeling of industrial beds it will be necessary to consider it.

Mathematically speaking, it is possible to neglect this term when the axial dispersion term is much smaller than the convective one.
This reduces to

\[ \frac{P_{e_a}}{D_a} = \frac{U_0 L}{D_a} \ll 1 \]

7) The cloud/wake and bubble concentrations are well mixed. This assumption is in agreement with the mass transfer models such as Chiba and Kobayashi's (1970, 1975) and Toei et al (1968, 1969). However, as discussed for condition (5), this should be a rather bad assumption unless the cloud-wake fraction is small.

II-3. THE FLUID MECHANICAL MODEL USED FOR DATA REDUCTION. Frequently, many of the parameters needed to determine the exchange coefficient are not measured but calculated from different fluid mechanical models. In an attempt to reconstruct the experiment it is necessary to derive fundamental parameters such as bubble diameters from a bed model. The model applied here is a simplified version of the one developed by Hughes (1977) whenever his model could not be directly applied. The modifications which were necessary were (a) the Cranfield and Geldart frequency (1974) correlation was usually not needed, as most experiments analyzed were performed in beds which were not freely bubbling; (b) the cloud was generally neglected as usually the bubbles were very fast. A model with these simplifications follows:

II-3.1 The Model. It is assumed that the cloud volume may be neglected or included as a part of the wake.

By conservation of mass, the total mass flow rate of gas across
any section of the bed will be constant and equal to the gas mass flow rate in the bubble and emulsion phases. Assuming that the gas is incompressible,

\[ F_T = F_b + F_e \quad \text{(II-2)} \]

where the \( F \)'s are the total bubble and particle gas volume flow rates at any cross section of the bed. The emulsion flow rate is

\[ F_e = A_T u_e \varepsilon_{mf} [1 - \delta(1 + f_w)] \quad \text{(II-3)} \]

where \( A_T \) is the bed cross section, \( u_e \) the interstitial velocity in the particulate phase. The bubble flow rate

\[ F_b = (1 + f_w \varepsilon_{mf}) \delta A_T u_b \quad \text{(II-4)} \]

The bubble and interstitial velocities

\[ u_b = u_o - u_{mf} + 0.711 (g \delta d_b)^{1/2} \quad \text{(II-5)} \]

and

\[ u_e = \frac{u_{mf} - f_w \delta u_b}{\varepsilon_{mf} \frac{1 - \delta (1 + f_w)}{1 - \delta (1 + f_w)}} \quad \text{(II-6)} \]

Eqns. (5) and (6) may be found in Kunii and Levenspiel (1968). If the beds superficial velocity \( u_o \) is known, then

\[ F_T = A_T u_o \quad \text{(II-7)} \]

Combining all the above equations, an equation with \( \delta \) and \( d_b \) as unknown variables and \( f_w \) as parameter is obtained.
\[ 0.711 g^{1/2} \delta + [ ( u_o - u_{mf} ) - ( 1 + f_w ) u_{mf} ] \delta - ( u_o - u_{mf} ) = 0 \quad \text{II-8} \]

\( \delta \) is then found in terms of \( d_b \), depending on the information given by the investigators. Davies and Richardson (1966) and Pereira and Calderbank (1975) submit bubble injection frequencies \( f_w \). For this case

\[ \delta = \frac{f_w V_b}{A_T u_b} \quad \text{II-9} \]

where \( V_b \) is the bubble volume. Eqn. (8) is then reduced to

\[ 0.3723 N^{1/2} \frac{f_w}{A_T} d_b^{1/2} + 0.5235 \frac{f_w}{A_T} [ ( u_o - u_{mf} ) - ( 1 + f_w ) u_{mf} ] \frac{f_w}{A_T} d_b^{2/3} - \\
\quad - 0.711 ( u_o - u_{mf} ) d_b^{1/2} - ( u_o - u_{mf} ) = 0 \]

Stephens et al (1967) submit a bubble density, \( N \), number of bubbles per unit volume, so

\[ \delta = N V_b \]

and eqn. (8) becomes

\[ 0.3723 N^{1/2} d_b^{1/2} + 0.5235 \frac{f_w}{A_T} N d_b^{2/3} - ( u_o - u_{mf} ) = 0 \]

These equations may then be solved for \( d_b \) once \( f_w \) is chosen. Usually this was taken to be either 0 or 1/3. Note that both equations give one unique positive solution for \( d_b \). Kunii and Levenspiel (1968), Fryer and Potter (1972) and Latham and Potter (1970) have purposed similar models. For the larger particle systems of Stephens' et al (1967) work, large clouds were encountered. Then \( f_w \) factor was used to include the cloud
fraction as part of $f_w$ by an iterative method.

For the experiments of Latham and Potter, Hughes' (1977) complete fluid mechanical model was used. In this case the Cranfield and Geis (1974) frequency versus height equation was used.

II-3.2 Discussion. Although the writer coincides with Yates (1975) that the bed hydrodynamics are of utmost importance in determining the bubble-emulsion mass exchange, a complete discussion of the model is not within the scope of this work. However, due to the important effect that the wake has in the determination of the experimental interchange coefficient, it will be analyzed in depth.

Wake. This region of the particulate phase has been included in the more recent bed models, such as the Kunii and Levenspiel (1969) bubbling bed model and the Latham, Hamilton and Potter (1968) and Fryer and Potter (1970) counter current back-mixing model, to explain the phenomenon of back-mixing, which was first reported by Gilliland and Mason (1949).

Part of the emulsion directly below the bubble has shown to accompany it in its rise by Rowe and Partridge (1962). To maintain continuity, the rest of the emulsion phase must descend.

Values for this parameter, which is commonly assumed a constant fraction of the bubble volume, has varied considerably. Partridge and Rowe (1965) made an x-ray study of rising bubbles and by assuming that the wake was the fraction of the sphere of which the void was the uppermost part, observed wake fractions of about $1/3$. Latham and Potter (1970) used a back-mixing technique and concluded that the bubbles were being followed
by an equal volume of the emulsion phase. Questions arise on whether all
the wake is in the defined sphere and whether a considerable fraction of
the emulsion is rising independently of the bubbles themselves. Both
groups of authors support their assumptions by quoting observations re-
ported by Rowe, Partridge, Lyall and Ardran (1962), and Woollard and Potter
(1968) respectively.

The variation in wake fraction and the lack of knowledge on its
dependence on different bed properties has an important effect on the
errors in the fluid mechanical behavior of the bed and especially on the
determination of the exchange coefficient. Most authors who account for
this region assume it to be at the same concentration as the cloud, al-
though it possibly is leaner as the gas within it is just returning to
the bubble. Also, the flow in this region is not understood properly as
there may be a large amount of convective mixing of the gases of both
phases when the exchange coefficient is determined experimentally, an
error arises because of the fluid mechanic inaccuracies due to our present
ignorance of the wake fraction. This is because the exchange coefficient
is proportional to the emulsion gas flow rate which becomes very sensitive
to the wake fraction in small diameter beds, typical for this type of ex-
periment.

II-4 THE MASS TRANSFER EQUATIONS

II-4.1 The Equations For data reduction in the frame of reference in which
the bubble is stationary, the bubble and cloud concentrations may be re-
lated by
\[ \frac{d N_b}{dt} = -K_1 (c_b - c_e) \tag{II-10} \]

where \( N_b \) is the number of moles of the gas of interest inside the bubble; 
\((c_b - c_c)\) is the mass transfer driving force assumed to vary only with time, 
and \( K_1 \) is the mass exchange coefficient.

A second equation describes the transfer from the cloud and wake to the bubble and emulsion phases.

\[ \frac{d N_c}{dt} = -K_1 (c_c - c_b) - K_2 (c_c - c_e) \tag{II-11} \]

\( N_c \) is the number of moles of gas within the cloud, \((c_c - c_b)\) and \((c_c - c_e)\) are the driving forces which are assumed not to vary in the neighborhood of the bubble except with time, and \( K_1 \) and \( K_2 \) are the standard mass exchange coefficients. \( c_c \) is an average cloud and wake concentration.

These equations may be transformed by noting that

\[ \frac{dt}{u_b} = \frac{dh}{V_b} \quad N_b = V_b c_b \quad \text{and} \quad N_c = \varepsilon_{mf} (V_c N_c + V_w) \]

\( V_b \), \( V_c \) and \( V_w \) being bubble cloud and wake volumes. The void fraction \( \varepsilon_{mf} \) within the cloud-wake region is assumed to be that of the emulsion.

Then,

\[ \frac{u_b}{V_b} \frac{d(V_b c_b)}{dh} = -\frac{K_1}{V_b} (c_b - c_c) \tag{II-12a} \]

and

\[ \frac{u_b \varepsilon_{mf}}{V_b} \frac{d(V_c + V_w)c}{dh} = -\frac{K_1}{V_b} (c_c - c_b) - \frac{K_2}{V_b} (c_c - c_e) \tag{II-12b} \]
Three different approximations to these equations have been examined.

II.4.1.1 *Model (1)* constant bubble and cloud/wake volume. The equations reduce to:

\[ u_b \frac{d C_b}{dh} = -K_{bc} (C_b - C_c) \]  
\[ u_b \varepsilon_{mf} (f_w + f_c) \frac{d C_c}{dh} = -K_{bc} (C_c - C_b) - K_{ce} (C_c - C_e) \]

where

\[ K_{bc} = \frac{K_1}{V_b}, \quad K_{ce} = \frac{K_1}{V_b} \]

and \( f_w \) and \( f_c \) are the wake and cloud to bubble volume fractions.

These equations are expected to be valid when there is no coalescence within the bed. This aspect is discussed later.

Fryer and Potter (1972, 1975) proposed these equations as part of their countercurrent back-mixing model. Besides the computational difficulties involved, the main disadvantage lies in that they are directly linked to the Kunii and Levenspiel interchange models as they are the only ones to have proposed this type of double resistance model. Subsequently, it will be called the modified Kunii and Levenspiel model.

II.4.1.2 *Model (2).* Either (a) the cloud/wake volume, or (b) the resistance between the cloud and emulsion or (c) that between the cloud and bubble is neglected. Then

\[ \frac{u_b}{V_b} \frac{d}{dh} \frac{V_b C_b}{C_b} = -K_{be} (C_b - C_e) \]
For the different assumptions, the meaning of the $K_{be}$ is given in Table (II-1)

### Table II-1

Different assumptions leading to Model 2.

<table>
<thead>
<tr>
<th>Property Neglected</th>
<th>$K_{be}$</th>
<th>$C_c$</th>
<th>Author</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) Volume of Cloud/wake</td>
<td>$1/K_{be} = 1/K_{bc} + 1/K_{ce}$</td>
<td></td>
<td>Kunii and Levenspiel ('68)</td>
</tr>
<tr>
<td>b) $K_{ce} = \infty$</td>
<td>$K_{be} = K_{bc}$</td>
<td>$C_c = C_c$</td>
<td>Davidson and Harrison ('63)</td>
</tr>
<tr>
<td>c) $K_{bc} = \infty$</td>
<td>$K_{be} = K_{ce}$</td>
<td>$C_c = C_c$</td>
<td>Toei and coworkers ('68, '69)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Partridge and Rowe ('66)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Chiba and Kobayashi (1970)</td>
</tr>
</tbody>
</table>

It should be noted that assumption (a) is only adequate for fast bubbles where $\alpha = u_b/u_f >> 1$. Considering that under these circumstances the models yield far higher values of $K_{bc}$ compared to $K_{ce}$, the resulting equation for the mass transfer coefficient is $K_{be} = K_{ce}$, which reduces case (a) to case (c).

The advantage of this form of Eqn. 14 is that growing bubbles may be treated adequately as bubbles in freely bubbling beds grow by coalescence. Unfortunately, as Pyle (1972) indicates, there is little confirmation of Toei and Matsuno's (1970) coalescence theory. This is
probably because of the difficulty of measuring accurately some of the parameters involved.

II-4.1.3 Model (3) Same assumptions as in (2) as well as constant bubble volume

\[
\frac{u_b}{dh} \frac{dC_b}{dh} = -K_{be} (C_b - C_e)
\]

II-15

The meaning of \(K_{be}\) and the value of \(C_c\) is as given in Table (II-1) for all the cases. The model has been used by many investigators in order to reduce their data, possibly because of its greater simplicity in manipulation.

Table (II-2) is a summary on which authors used which models, some slightly modified.

Table (II-2)

Models Used by Different Authors for Reduction of Experimental Data

<table>
<thead>
<tr>
<th>Model 1</th>
<th>Model 2</th>
<th>Model 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fryer &amp; Potter (1975, 1976)</td>
<td>Chavarie &amp; Grace (72, 75, 76)</td>
<td>Chavarie &amp; Grace (72)</td>
</tr>
<tr>
<td>Rowe, Evans &amp; Middleton (71)</td>
<td>Drinkenburg &amp; Rietema (1973)</td>
<td>Drinkenburg &amp; Rietema (1973)</td>
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<td></td>
<td>Davies &amp; Richardson (1966)</td>
<td>Davies &amp; Richardson (1966)</td>
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</table>
II-4.2 Discussion

1) Bubble volume. The assumption that the bubble volume is constant is generally considered good when bubbles are not coalescing and there is no excess gas within the emulsion phase itself. Goldsmith (1975) and Pereira and Calderbank (1975) confirm that single bubble properties do not vary with height, but with diameter. However, Rowe et al (1971) found seemingly identical bubbles traveling at noticeably different velocities. In a great number of papers presented single bubbles were injected into the bed which, after formation, were seen to remain at a constant volume. In other cases, such as Stephens et al (1967) and in all of the cases of freely bubbling beds (see Table (III-2)) growth of the bubbles by coalescence was expected but not accounted for. For the latter it is possible to reprocess the raw data by using a frequency correlation like Cranfield & Geldart's (1974) to determine bubble growth. However, in the former cases this is not possible and a constant bubble volume must be assumed.

As Chavarie and Grace (1976) indicate when a tracer bubble is injected and it grows inside the bed, its concentration becomes diluted because of the influx of gas which may mistakenly be taken as a reduction of tracer due to exchange. Consequently bubble concentration profile measurements may yield artificially high exchange coefficients when the bubble growth is neglected. In their experiments they differed by a factor of 2.5. Note that the bubble growth observed by them was due to operation at excessive gas flow rates in the emulsion phase.

It should be remarked that although bubble growth has been
allowed for in the general model, a step change in the volume of the bubble itself by coalescence hardly will be represented by the exchange coefficients \( K_1, K_2 \) studied the concentration differences between the phases. In Appendix (II-3) an alternative form to account for coalescence is given which assumes the bubble changing volume continuously with height. Toor and Calderbank (1967) have claimed that this process is unimportant but many others, such as Chiba and Kobayashi (1970) and Pereira and Calderbank (1975) do indicate that exchange in a freely bubbling bed must be considerably higher. The latter specify that the increase must be by this mechanism.

2) Cloud. In many of the experiments made, the bubble is very fast so the cloud is small. However the wake has generally been found to be a sizeable fraction of the bubble and so the cloud/wake fraction is always considerable. Hence model (2a) is questionable. The danger of this simplification is seen by comparing the models represented by Eqns (13) and (15) as done in Figures III-7 and 8 for data from Stephens et al (1967).

3) Boundary Conditions. For the first two models the boundary conditions are essential to the resultant shape of the concentration curves. These are usually taken at the bottom of the bed where frequently there are large disturbances as Stephens et al (1967) and Chavarie and Grace ('75, '76_ have indicated. Consequently, one interchange model may be favored over another because of their sensitivity to the boundary conditions. On the other hand, Model 3 presented the comparison which may be
reduced to finding the slope of a modified logarithmic concentration profile, becoming independent of such boundary condition. Similar methods could be used for Model 1 by the methods shown by Douglas (1972). However, because of the scatter in the data large errors would be incurred upon.

The boundary conditions used will be deduced here. In the case that bubbles are formed by injecting tracer gas in a non-freely bubbling bed, it may be assumed that the bubbles instantly acquire the dimensions of the average bubble within the bed, as well as a cloud and a wake. The gas injected is used to occupy the bubble and, if there is an excess the rest is diluted to form the cloud. If the assumptions used require the gas in the bubble/cloud/wake regions to be uniform, the tracer is averaged out. Note that at this stage none of the gas injected enters in the emulsion phase if the bed is minimally fluidized as all excess gas must go in the form of bubble/cloud/wake even for the simpler two phase flow models. These boundary conditions are commonly used in the literature (Walker, 1975, and Rowe et al, 1971 are the most explicit). They are quite artificial. Discussions on their validity are given by Rowe et al (1971) and Chavarie and Grace (1975).

In the case of back-mixing, that is when the emulsion velocity is downwards, other boundary conditions must be used, such as those of Fryer and Potter (1972).

The equations may be integrated once the boundary conditions have been decided upon. These are found in Appendix (II-4). The simplicity of the form of the third model show why it has been the most
popular. As the slope of the logarithmic concentration profile is proportional to the interchange coefficient, the result is independent of the boundary conditions. This is in contrast with the other more sophisticated models which depend importantly on them. A final note of warning must be given about the simpler model: because the slope has to be taken from a semilog plot and the argument of the logarithm approaches zero, there may be a large scatter in the data points plotted.

II-5 THE INTERCHANGE MODELS

There are a large number of gas bubble to emulsion interchange models. Chavarie and Grace (1976) have listed 13, not all of them having been fully described in the available literature yet. It is the author's intention only to describe the more widely applied interchange coefficients, analyze the different methods used to account for them and compare these with the methods used to explain other mass transfer problems.

Following Drinkenburg and Rietema (1972), the different models will be classified depending on where the author considers the highest resistance to mass transfer between the bubble and emulsion phases lies. These resistances are found in the following regions:

a) at the bubble-cloud interface on the bubble side

b) at the bubble-cloud interface on the cloud side

c) through the cloud thickness

d) at the cloud-emulsion interface on the cloud side

e) at the cloud-emulsion interface on the emulsion side

Fig. 2 illustrates the locations of these resistances.
Figure II-2 Location of the Different Mass Transfer Resistances
The mechanisms considered are either due to convection or molecular diffusion. The bubbles are assumed to be well mixed as well as the emulsion in the neighborhood of the bubble except for a film at the interfaces in the case of the diffusion models.

Figure 3 summarizes the location of the resistances that have been accounted by some of the models. The equations pertaining to each one is included in Table (II-3). Except for three convective mechanisms, the mechanisms which are derived are all very similar and will be discussed collectively.

These models suggest either (a) a convective and/or (b) a diffusive mechanism for mass transfer.

(a) Convection. These mechanisms are of two types: those occurring at the bubble–cloud interface and those occurring at the cloud–emulsion interface. The former assume that all the gas within the bubble is in contact with the particulate phase, the bubble and particulate phases being at different concentrations. There is a renewal of gas within the bubble. The throughflow may be up to \( 3 \, u_{mf} \left( \frac{\pi d^2}{4} b \right) \) for the Davidson and Harrison and Kunii and Levenspiel models, while it has a reduced value of \( 1.90 \, u_{mf} \left( \frac{\pi d^2}{4} b/4 \right) \) in the model by Walker. In all cases the throughflow is a consequence of the bubble model used (Davidson and Harrison's, Murray's etc.). It should be noted that the only difference between the Hovmand & Walker models is that one is based on the Davidson and Harrison bubble model while the other on Murray's.
Figure II-3 Location of the Different Mass Transfer Resistances According to Different Authors

<table>
<thead>
<tr>
<th></th>
<th>bubble</th>
<th>a</th>
<th>b</th>
<th>cloud</th>
<th>a</th>
<th>c</th>
<th>d</th>
<th>e</th>
<th>emulsion</th>
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<tbody>
<tr>
<td>Chiba and Kobayashi (1970)</td>
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<td>Davidson and Harrison (1963)</td>
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<td>Drinkenburg and Rietema</td>
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<td>Normand (1968)</td>
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<td>Kato &amp; Wen (1969)</td>
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<tr>
<td>Kunii &amp; Levenspiel (1968)</td>
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<tr>
<td>Partridge and Rowe (1966)</td>
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<tr>
<td>Rowe (1964)</td>
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<tr>
<td>Toel and Matsuno (1968)</td>
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<tr>
<td>Walker (1975)</td>
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</table>

Key: C - Convection; D - Diffusion; A - Adsorption to particles; E - Empirical; N - No Exchange; gas is well mixed
### Different Mass Transfer Equations

**Table (II-3)**

<table>
<thead>
<tr>
<th>Source</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Davidson and Harrison</td>
<td>$K_{be} = 4.5 \frac{u_{mf}}{d_B} + 5.85 \left( \frac{D_b g}{D_b \frac{1}{2}} \right)^{1/2}$</td>
</tr>
<tr>
<td>Toel et al*</td>
<td>$K_{be} = 4 \frac{\varepsilon_{mf}}{1 + 2 \varepsilon_{mf/}(\alpha-1)} \frac{\lambda A_{out}}{n d_B} + 4.08 \frac{\sqrt{u_b D/\alpha^2-1}}{d_B^3}$</td>
</tr>
<tr>
<td>Chiba and Kobayashi</td>
<td>$K_{be} = \left[ 1 + \frac{2}{3} \frac{m}{\varepsilon_{mf}} \frac{1-\varepsilon_{mf}}{2} \left( \frac{1 + \alpha}{\alpha-1} \right) \right]^{1/2} \times 6.78 \frac{D \varepsilon_{mf}^2 u_b}{1 - f_w} \left( \frac{D_e \varepsilon_{mf} u_b}{D} \right)^{1/2}$</td>
</tr>
<tr>
<td>Kunii and Levenspiel</td>
<td>$K_{be} = 4.5 \frac{u_{mf}}{d_B} + 5.85 \left( \frac{D_b g}{D_b \frac{3}{2}} \right)^{1/2}$ $\frac{1}{K_{ce}} = \frac{1}{K_{be}} + \frac{1}{K_{bc}}$ $K_{ce} = 6.78 \left( \frac{D_e \varepsilon_{mf} u_b}{D_b^3} \right)^{1/2}$</td>
</tr>
<tr>
<td>Partridge and Rowe</td>
<td>$K_{be} = \frac{\varepsilon_{mf} D}{n d_B^2} \left( 2 + 0.69 \left( \frac{V_D}{D} \right)^{1/3} \right) \frac{\left( \frac{u_b - u_f}{V} \right) \left( \frac{6Vc}{\pi} \right)^{1/3}}{1/2}$</td>
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</table>
### Table (II-3) (Continued)

<table>
<thead>
<tr>
<th>Source</th>
<th>Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hovmand</td>
<td>$K_{be} = \frac{7.14 \frac{F}{u}}{\frac{F}{u} + \epsilon_{mf} \frac{F}{u}} \left[ u_{mf} + 0.766 \frac{F}{u} \epsilon_{mf} \left( \frac{D_{a} g}{d_{b}^{1/2}} \right)^{1/2} \right]$</td>
</tr>
<tr>
<td>Walker</td>
<td>$K_{be} = \frac{2.83 \frac{F}{b}^{1/2}}{\frac{F}{b}^{1/2} + \epsilon_{mf}^{1/2}} \left[ u_{mf} + 1.93 \frac{F}{b}^{1/2} \epsilon_{mf} \left( \frac{D_{a} g}{d_{b}^{2/2}} \right)^{1/2} \right]$</td>
</tr>
<tr>
<td>Drinkenburg and Rietena</td>
<td>Numerical</td>
</tr>
<tr>
<td>Kato and Wen</td>
<td>$K_{be} = \frac{11}{d_{b}}$</td>
</tr>
<tr>
<td>Rowe</td>
<td>$K_{be} = 0$</td>
</tr>
</tbody>
</table>

*For two dimensional beds*
Two interesting forms of convection are described by the models of Toei et al (1968, 1970). The first, initially suggested qualitatively by Rowe et al (1963) and later by Davies and Richardson (1967) consisted of wake shedding. Tracer gas experiments have been made showing that tracer is left behind the bubble in the pattern of a Christmas tree. Rowe explains this as a method of shedding vortices, one to each side. The interchange is calculable if the volume of the gas lost and the frequency of the shedding were known. Unfortunately these parameters are not easily measured in a fluidized bed.

The other form of convective interchange suggested by Toei et al (1970) describes a mechanism of gas loss due to coalescence of bubbles. The description is that of Pyle (1972). In order to maintain a given bubble flow rate within the bed, as required by the two phase model, two bubbles that coalesce to form a third must lose some gas as the velocity of the latter is greater. As Pyle (1972) indicates, there has been little support for Toei's work.

Chiba and Kobayashi (1970,75) include two terms which would account for mass transfer by gas absorption to the particles as they go from the cloud to the emulsion. However, as in the case of Toei et al, in the author's knowledge this part of these models has yet to be independently verified. Chiba and Kobayashi (1970) do recognize that there is scarce information on the absorption factor m in use. They do claim that the effect should be relevant. However, experiments by Drinkenburg and
Rietema (1973) by using different tracer gases and of Chavarie and Grace (1976) by using particles with different absorption characteristics have led to the conclusion that absorption is unimportant.

b) Diffusion. All the diffusion mechanisms assume the existence of a well defined streamline surface at the interface of interest (bubble-cloud or cloud-emulsion) which separates the flow at either side. In this situation the resistance to mass transfer is molecular diffusion.

Partridge and Rowe (1966) compare the bubble-cloud and emulsion to a system consisting of an immiscible liquid rising in another and then suggest the use of Rowe et al's (1965) correlation for mass transfer from a solid sphere to a fluid.

Kunii and Levenspiel at the cloud-emulsion interface apply the penetration theory of Higbie (1935). (This is Stokes first problem of viscous penetration - see Schlichting (1968) - applied to diffusion.) The method was originally applied to gas-liquid interfaces where the contact time is short. In fact, the theoretical value of the mass transfer coefficient by this method for a drop in a gas or a gas bubble in a liquid has an identical solution, after taking into account the change in the molecular diffusion coefficient due to the porosity of the emulsion phase. Sherwood et al (1976) note that for the gas-liquid the coefficients has, to be reduced for greater accuracy.

The other method of calculation of the diffusion term was originally proposed by Boussinesq (1906) for heat transfer to an inviscid fluid from a solid surface. It consists of writing the diffusion equa-
tion using the stream and potential functions as the basis for the axes. The solution to the above problem of a drop in a gas gives by this method an identical answer to the Higbie penetration method. This is the reason why the Chiba and Kobayashi diffusion term is the same as the Kunii and Levenspiel one, except for a factor of $\varepsilon \frac{mf}{1-f_w}$, which enters due to a change in volume and a different interpretation of the equivalent molecular diffusion $D_e$. The authors found it necessary to approximate the cloud-emulsion interface as a sphere. However, the identical result is obtained for the Davidson and Harrison bubble model exactly. This model applied the streamfunction of Murray's spherical bubble.

Toei et al (1968) had earlier proceeded in the same fashion as Chiba and Kobayashi (1970) but by applying Davidson and Harrison's two dimensional streamfunction. Also, a volumetric correction factor included slightly different considerations.

Davidson and Harrison (1963) use the earlier results of Baird and Harrison (1962) for diffusion from a spherical cap gas bubble in a liquid. The method used is, again, Boussinesq's. There is an obvious inconsistency in the two mechanisms (flow through the bubble/cloud interface and a diffusive resistance due to flow along the interface) because if all the gas goes through the void, the streamlines cannot be parallel to the bubble-cloud interface, and consequently may not be superposed as convection is high. Pyle and Rose's (1965) work suggests a reconciliation by showing that some gas recirculates within the bubble while some others flow through it, however the streamlines are not parallel to the
interface. So, from the theoretical standpoint, the Davidson and Harrison interchange model should be an overestimation. Hovmand (1968) derives the exchange resistance on the bubble-cloud interface in the same way, but decreases the amount of throughflow. The analysis for the bubble-cloud interface, on the bubble side, is apparently that of Van Heusen and Beek (1968) for a gas slug in a liquid. Hovmand adds to this the resistance at the same interface but on the cloud side. Reportedly Walker (1975) does the same but uses Murray's potential flow model.

   c) Other models. Drinkenburg and Rietema (1972) proposed a model which is developed from a linearized form of the diffusion equation, which accounts for gas absorption to particles, with the aid of either Davidson and Harrison's (1963) or Murray's streamfunction. In this way they are able to account for all the resistances to mass transfer within the particulate phase. The model is mathematically involved (it must be solved numerically) and uses some parameters which are not well known such as an effective diffusion coefficient within the particulate phase and the porosity of the particles. A great disadvantage of this model is that it does not offer a closed solution. This is probably why the only experimental work published up to now that has attempted to check the model belongs to the proposers themselves.

Kato and Wen (1969) adopted for their bubble assemblage model an empirical interchange coefficient proposed by Kobayashi, Arai and Chiba (1965). However, in a later paper, Chiba and Kobayashi (1975) claim that the correlation is not valid in fine particle systems.
Rowe (1964) initially proposed a bubble model where bubble and cloud were assumed to be well mixed and the resistance between the bubble/cloud and emulsion was very large, making mass transfer negligible.

From the above discussion certain general tendencies of the models may be expected. If the general behavior of the bubble models is accepted as adequate, it may be anticipated that the models relying primarily on convection from the bubble to the emulsion without any cloud residence should overestimate the mass transfer while those relying exclusively on molecular diffusion from the cloud to the emulsion should underestimate it. The first neglect that the gas recirculates around the bubble void, while the latter rely too heavily on a well defined streamlined surface.

In the literature it is generally agreed that the Rowe and Partridge theory gives consistently low results (see, for example Pyle (1972)), and that the Davidson and Harrison is either adequate or overestimates (see Walker (1975)). Of the models the outstanding one is that by Kunii and Levenspiel, which some authors feel has best approximated their results (Grace and Chavarie (1975), Fryer and Potter (1976)). However, many reviewers such as Pyle (1972), Yates (1975), Rowe (1972) and of a number of more recent experimentors (Chavarie and Grace (1975, 1976), Walker (1975), Rietema and Hoebink (1975) believe that there is no satisfactory model.

It was decided to compare only three models in detail to data available. The models selected were those of Davidson and Harrison (1963),
Kunii and Levenspiel (1968) and Chiba and Kobayashi (1970). In the latter
gas absorption to particles was neglected. It was felt that they were
representative as they considered different resistance mechanisms, were
commonly used and required the knowledge of parameters which are usually
reported with the experiment.

II-6 **SUMMARY.** Equations for the reduction of data and the theoretical
bubble to emulsion mass transfer models have been presented and discussed
with many references to investigators in the field. It is apparent that
the models are still at a primitive stage.
II-7 NOMENCLATURE FOR CHAPTER II AND ITS APPENDICES

$A_T$ - total bed cross section [$L^2$]

$A_{out}$ - bubble area for Toei and Matsumo's (1968) correlation [$L^2$]

$C_a$ - tracer concentration on the axis [-]

$C_b, C_c, C_e$ - tracer concentration in bubble, cloud and emulsion [-]

$C_{co}$ - tracer concentration at cloud emulsion interface [-]

$C_{\infty}$ - volumetric average tracer concentration [-]

$C_o$ - tracer concentration at the injection point [-]

$\Delta c$ - emulsion tracer concentration at the bottom and top of bed [-]

$D$ - molecular diffusion coefficient [$L^2/T$]

$D_e, D_a, D_r$ - effective and effective axial and radial dispersion coefficients [$L^2/T$]

$d_b$ - effective bubble diameter [L]

$F_b, F_c, F_e, F_T$ - bubble, cloud, emulsion and total flow rates [$L^3/T$]

$F_o$ - tracer injection flow rate [$L^3/T$]

$f_c, f_w$ - cloud and wake to bubble volume fraction [-]

$f, f_\ell$ - bubble point and level frequencies [l/T]

$g$ - gravitational acceleration [L/T]

$h$ - height, vertical coordinate within the bed [L]

$k_1, k_2$ - bubble to cloud and cloud to emulsion mass transfer coefficients [$L^3/T$]

$k_{bc}, k_{ce}, k_{be}$ - bubble to cloud, cloud to emulsion and bubble to emulsion mass transfer coefficients, per unit bubble volume [l/T]
L - height of bed [L]

m - particle adsorption coefficient [-]

N - number of bubbles per unit bed volume [1/L^3]

N_b, N_c - tracer contained in bubble and cloud [L^3]

Pe_a - effective axial Peclet = u_o L/D_a [-]

R - radius of bed [L]

r_b - effective bubble radius [L]

r_c - effective cloud radius [L]

S(r,z) - source [L/T]

t - time variable [T]

u_b, u_br - bubble velocity and velocity of bubble in a stagnant bed [L/T]

u_mf, u_f - superficial and interstitial velocities in a minimally fluidized bed [L/T]

u_o - superficial velocity in a fluidized bed [L/T]

V_b, V_c, V_w - bubble, cloud and wake volumes [L^3]

Greek Symbols

α - u_br/u_f [-]

δ - bubble to bed volumetric fraction [-]

ε_mf - bed voidage at minimum fluidization [-]

θ - polar angle [-]

φ - potential function [L^2/T]

φ' - modified potential function = r^2 sin^2 δ dφ [L^4/T]

ψ - stream function [L^2/T]
REFERENCES TO CHAPTER II AND ITS APPENDICES

REFERENCES TO CHAPTER II AND ITS APPENDICES (Cont.)


REFERENCES TO CHAPTER II AND ITS APPENDICES (Cont.)

CHAPTER III

STUDY OF THE GAS MASS TRANSFER
FROM FAST BUBBLES TO EMULSION PHASE II

Analysis of Experimental Work and
Comparison to Theoretical Models

III-1 Introduction

Twenty papers have been found which present data on mass transfer from bubble to emulsion. However, only four of them were judged adequate for detailed analysis, with aid of the equations presented in the last chapter. Three of the papers studied bubble to emulsion mass transfer from bubbles injected into a minimally fluidized bed and the other to a freely bubbling bed with backmixing. Only in one of the experiments were both emulsion and bubble concentration measured simultaneously. The data was compared to the theoretical exchange models of Davidson and Harrison, Kunii and Levenspiel and Chiba and Kobayashi. None of the models is definitely favored over the other, but some trends are apparent. In the final section all the data published on mass exchange from bubble to emulsion in minimally fluidized beds is united in a single graph. Again, certain trends are found.

III-2 VALIDITY OF EXPERIMENTAL WORK

As indicated by Rietema and Hoebink (1975) a standard method of determining the exchange coefficient experimentally is to inject bubbles rich in a tracer gas into a minimally fluidized bed. The tracer concentration is then measured throughout the bed, either in the bubble or emulsion phase or in both. The drawback of this method is that the conditions imposed are
not those found in a freely bubbling bed. The behavior of swarms of bubbles is generally considered to be different to that of single bubbles as Calderbank et al (1975), Chavarie and Grace (1975) and Chiba and Kobayashi (1970) have commented. However, the models proposed refer to single bubbles with exception to Toei and Matsuno's (1970). Another problem, is the difficulty of injecting gas into bubbles only. In fact, measurements at the bottom of the bed commonly appear to be erratic as may well be the case in Stephens et al (1967) and Davies and Richardson (1966) work. Rowe et al (1971) describe effects at the level close to the bottom of the bed.

Another method used originally by Gilliland and Mason (1949) and more recently by Latham and Potter (1970) and Fryer and Potter (1976), consists of injecting tracer into the emulsion phase close to the top of the bed. The bed is made to bubble at high enough a superficial velocity that the gas in the emulsion phase starts to descend. Then the tracer concentration within the bubbles will increase as they rise. Although this is a more realistic representation of a fluidized bed, it will be seen that there are some difficulties encountered particularly due to the lack of definition of the wake fraction, which is the driving mechanism for back-mixing.

As there is only scarce knowledge of the detailed behavior of a fluidized bed, the methods used to determine the interchange coefficient must be as simple and direct as possible. Many of the experiments will be dismissed for any of the following reasons.
(i) Interchange from slugging beds. It is generally accepted (e.g., Walker, 1975, Pyle, 1972, and Calderbank et al, 1975) that the mass exchange is different for this case. This is quite obvious as the behavior of a slug such as its velocity is quite different to that of a bubble. In general, results for slugs such as those of Pereira and Calderbank (1975) and, according to Pyle (1972), suggest that the interchange is high for this case and well represented by the Davidson and Harrison (1963) or Mowmand and Davidson (1968) models. This is because the cloud is essentially in contact with the walls of the bed and then swept away.

(ii) Two dimensional beds. Although there are a number of good investigations in two dimensional beds, it is thought that they really do not represent ideal two dimensional behavior, as Rowe (1971) indicates. This may primarily be due to wall effects. Chavarie and Grace (1975) and Rowe (1971) recognize the inapplicability of their two dimensional results to the three dimensional case. Walker (1975) indicates that in his two dimensional bed solids rained along the walls. This implies that a sort of plug flow motion is achieved in this dimension, which would increase the exchange. Walker (1975) found that the interchange was higher compared to theory than he found for the three-dimensional work. Due to the difficulties and dubiousness of the re-interpretation of the data, these experiments will be neglected.

(iii) Chemical reaction. A common method of studying freely bubbling beds is to inject a tracer which reacts with the particles. In this way measurable gradients in the vertical direction of the tracer are produced. Unfortunately, this produces local gradients in the cloud and emulsion phases
**Experiments of bubble to emulsion mass transfer**

**Table III-1**

<table>
<thead>
<tr>
<th>Study</th>
<th>Bed Dimension</th>
<th>Chemical Reactions</th>
<th>Slugs</th>
<th>Freely Bubbling</th>
<th>Sufficient Information</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chavarie and Grace (1972)</td>
<td>2</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Chavarie and Grace (1975)</td>
<td>2</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Chavarie and Grace (1975a, 1976)</td>
<td>2</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Chiba and Kobayashi (1970)</td>
<td>3</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Davies and Richardson (1966)</td>
<td>3</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Drikenburg and Rietema (1973)</td>
<td>2 &amp; 3</td>
<td>No</td>
<td>Some</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Fryer and Potter (1976)</td>
<td>3</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Guedes de Cravahlo &amp; Harrison (1975)</td>
<td>3</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Hovmand and Davidson (1968)</td>
<td>3</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Kobayashi, Arai and Sunakawa (1967)</td>
<td>3</td>
<td>No</td>
<td>Some</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Latham and Potter (1970)</td>
<td>3</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Overcashier, Todd and Olney (1959)</td>
<td>3</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Pereira and Calderbank (1975) (also, Calderbank et al 1975)</td>
<td>3</td>
<td>No</td>
<td>No</td>
<td>Some</td>
<td>Yes</td>
</tr>
<tr>
<td>Rietema and Hoebink (1975)</td>
<td>3</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Bed Dimension</td>
<td>Chemical Reactions</td>
<td>Slugs</td>
<td>Freely Bubbling</td>
<td>Sufficient Information</td>
<td></td>
</tr>
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<td>-------</td>
<td>-----------------</td>
<td>------------------------</td>
<td></td>
</tr>
<tr>
<td>Rowe, Evans, and Middleton (1971)</td>
<td>2</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Toei, Matsuno (1968) (also Toei et al (1969))</td>
<td>2</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Toei, Matsuno and Nishitani (1970)</td>
<td>2</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Stephens, Sinclair and Potter (1967)</td>
<td>3</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Szekely (1962)</td>
<td>3</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Walker (1975)</td>
<td>2 &amp; 3</td>
<td>Yes</td>
<td>Some</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>
and consequently resulting calculations are inaccurate, as Chavarie and Grace (1972) report. Other problems arise due to lack of knowledge of the chemical rate of reaction. Walker (1975) recognized that small errors in the reaction rate constant produced a large scatter in the exchange coefficient calculations. All these difficulties result in obscuring the value of the observed mass exchange itself, and hence these experiments must also be neglected.

Other problems arise mainly due to insufficient information on the raw data of the experiments being published, such as no concentration profiles, void fraction, bubble frequency, etc. Table (III-1) summarizes the works considered, and shows which papers share these characteristics.

III-3 PARTICULAR WORKS ANALYZED

Of all the experiments reported in Table (III-1), there are only four which seemed fit for detailed analysis. These are the early works of Davies and Richardson (1966) and Stephens et al (1967), with beds at incipient fluidization the freely bubbling bed results of Latham and Potter (1970) and the more recent and complete work of Pereira and Calderbank (1975) also at minimum fluidization. Average particle size $d_p$ and minimum fluidization velocity $u_{mf}$ are given in Table (III-2) for all of the experiments studied in detail. The experiments will be discussed separately and then compared using several non-dimensional forms.
<table>
<thead>
<tr>
<th>Investigator</th>
<th>$d_p$ ($\mu$)</th>
<th>$\epsilon_{mf}$</th>
<th>$u_{mf}$ (cm/s)</th>
<th>shape</th>
<th>type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Davies &amp; Richardson</td>
<td>55</td>
<td>0.5266</td>
<td>0.142</td>
<td>irregular</td>
<td>Cracker catalyst</td>
</tr>
<tr>
<td>Stephens, Sinclair &amp; Potter</td>
<td>590</td>
<td>0.39*</td>
<td>25</td>
<td>spherical</td>
<td>Ballotini</td>
</tr>
<tr>
<td></td>
<td>363</td>
<td>0.33*</td>
<td>11.2</td>
<td>spherical</td>
<td>Ballotini</td>
</tr>
<tr>
<td></td>
<td>290</td>
<td>0.39*</td>
<td>3.52</td>
<td>spherical</td>
<td>Ballotini</td>
</tr>
<tr>
<td></td>
<td>64</td>
<td>0.44*</td>
<td>0.39</td>
<td>spherical</td>
<td>Ballotini</td>
</tr>
<tr>
<td>Pereira &amp; Calderbank (1975)</td>
<td>30-105</td>
<td>0.557</td>
<td>0.575</td>
<td>irregular</td>
<td>partially graphitized</td>
</tr>
<tr>
<td>Latham &amp; Potter (1970)</td>
<td>97</td>
<td>0.44</td>
<td>1.12</td>
<td>spherical</td>
<td>Ballotini</td>
</tr>
</tbody>
</table>

*setttled bed voidage
III-4 PROCEDURE FOR CALCULATION OF THE EXCHANGE COEFFICIENT

III-4.1 Procedure for Davies and Richardson (1966) and Stephens et al's (1967) data. If the cloud fraction is small, it may be possible to describe the exchange process with the mass transfer model (3) for data reduction represented by equation (II-15), where the cloud/wake is either neglected or assumed to be at a given concentration (that of the emulsion or bubble). The bubble volume may be assumed constant. The solution is given in Appendix II-3.

Under these conditions, the experimental exchange coefficient may be calculated. The procedure is as follows.

a) adopt a fluid mechanical model to define bubble diameter, velocity and flow rates of the different phases. The model of Section (II-3) was used, where the wake fraction was a parameter.

b) find a least square fit of the concentration profile if the two phases were perfectly mixed. The fit is of the form \( \ln(C-C_\infty) = a + bh \) where \( C \) is the concentration, \( h \) the distance from the injector and \( a \) and \( b \) the coefficients to be determined.

c) calculate the values for \( K_{be} \) for the different bubble mass transfer theories and the information obtained from part (a) and the paper.

Some of Stephens et al's (1967) data could not be processed in this way. When the cloud fraction became large, it was more convenient to compare the theoretical concentration profiles with the data. The procedure was:
a') same as (a) for the previous case. However, in order to account for the cloud, the model developed by Hughes was used with a modified equation for bubble void fraction:

$$\delta = NV_B$$

where $N$ is the number of bubbles per unit volume reported by the authors.

b') with the information supplied in (a') and Table II-3 for the theoretical exchange coefficients, the theoretical concentration profiles are calculated using the mass transfer for data reduction models (1) and (3). As the clouds were big and an average concentration was reported it was assumed that the measured concentration was

$$C_{ave} = \delta(f_w + f_c) C_c + [1 - \delta(f_w + f_c)] C_e$$

and suggested by Fryer and Potter (1972).

For the models assuming a single exchange coefficient (Model 3), the term accounting for the concentration in the cloud was neglected as the theoretical bubble to emulsion exchange models neglect them.

The present procedures for data reduction differ with those used by the authors primarily in the fluid mechanical bed model. They assumed that all the gas injected in their incipiently fluidized beds formed bubbles, with no contribution from the gas originally in the emulsion phase. However, observing that the interstitial gas may not rise faster than $u_f$ (the emulsion gas velocity at minimum fluidization) and that the effective cross section for the emulsion phase of the bed at any height has on the average been reduced due to the presence of the bubbles, it is clear that the total gas
Flow Rates in (a) minimally fluidized beds; (b) and (c) bubbling beds; (b) and (c) represent the same conditions.

Figure III-1
flux through the emulsion phase must be less than $u_{mf}$. This is shown in Figure 1, where in Figure 1C all bubbles have been lumped together in the way of Fryer and Potter (1972). Consequently, some of the gas which was being used to minimally fluidize the bed must contribute to the bubble void or cloud/wake volume. So it is inadequate to assume that the injected gas is used exclusively to form the bubble. Richardson and Davies (1963) observed bubbles of larger volume than the volume of gas injected. On the other hand, Pereira and Calderbank (1975) measured smaller bubbles than the gas injected into the bed, possibly indicating that both injected gas contributed to form the cloud.

Stephens et al (1967) did not account for the influence of the large clouds in their experiments with the larger particles as has been done here.


It was possible to evaluate the interchange coefficient in a number of ways, by directly using the simple equation II-15 of Model (1) from which the coefficient for the constant volume, single resistance case may be found.

The equations used to make the evaluation is found in Table III-3. Also average values for the tracer flow rate and all the exchange coefficients the reported exchange coefficient and all the standard deviations are given. It should be noted that the variation between the average values may be large, as well as their standard deviation. This is not unexpected as the equations dealt with are of the finite difference type. $u_{b}$ and $Δh$ are measured quantities
Different methods of data reduction
Table III-3

\[ K_{be} = \frac{-U_b \frac{dC_b}{dh}}{C_b - C_e} \]

\[ \frac{-U_b \frac{C_b(h+\Delta h) - C_b(h)}{\Delta h}}{C_b - C_e} \]

\[ \frac{-U_b \frac{C_b(h) - C_b(h-\Delta h)}{\Delta h}}{C_b - C_e} \]

\[ \frac{-U_b \frac{C_b(h+\Delta h) - C_b(h)}{2\Delta h}}{C_b - C_e} \]

The above procedure is different to that published by the authors in that they used the slope of the straight line fitting the data on a graph with axes height (h) and logarithmic driving force \(2\ln(C_b - C_e)\). The slope is proportional to the exchange coefficient.

III-4.3 Procedure for Reducing Latham and Potter's (1970) Raw Data

In this paper the bed used was freely bubbling. In order to account for coalescence, it was necessary to account for variations in bubble diameters and consequently it was not possible to reduce the data to a mass transfer coefficient as a function of bubble diameter, as in most of the cases already considered in the last two paragraphs. It was preferred to compare the experimental concentration profile with the theoretical ones. The procedure was:

a) With data from the paper and use of Hughes' (1977) fluid mechanical model, the bubble diameter, velocity and cloud and the interstitial gas velocity in the emulsion were calculated as a function of height.
b) The theoretical mass transfer exchange coefficients of Davidson and Harrison, Kunii and Levenspiel and Chiba and Kobayashi in Table (II-3) were then inserted into the mass transfer equations for data reduction given by Model (1) and Model (3) (single resistance between bubble and emulsion allowing for varying and fixed bubble volumes, respectively). The bubble diameter has been defined as a function of height in part (a), and so all properties needed to solve the differential equations II-13 and II-14 are functions of bubble diameter. The solution to these equations is given by equation (2) of Appendix II-3 as an integral. This is then numerically integrated to obtain the emulsion concentration profile which was compared to the measured data. The contribution to the sampled concentration by the bubble void was neglected as the void fraction $S < 0.05$.

The authors did use in this case a bed model which was similar to the one used here for their data reduction. The difference lies in that they assumed that the bubble volume did not vary with height. The slope of the logarithmic concentration versus height curve could then be used to calculate the exchange coefficient for the different bubble diameters.

III-5 ANALYSIS OF THE PAPERS

III-5.1 Davies and Richardson (1966). Single bubbles of carbon dioxide - with molecular diffusivity of 0.146 cm$^2$/s according to Davies and Richardson (1967) are injected into the bottom of an incipiently fluidized bed 22.36 cms in diameter. The bubbles were injected at a rate of 44 per minute (0.73/sec) and it was assumed that all the tracer was used to form the bubble itself. A typical two phase model assumption. The bubbles were very fast and, consequently, the clouds were very thin (less than 1% of bubble diameter by the Davidson and Harrison (1963) bubble model criterion).
The tracer was sampled about 3.3 cms away from the wall in order to be outside the path of the bubbles. It was only measured at one position at each height after having concluded from a set of measurements at one height that radial concentrations within the bed were perfectly flat.

Unfortunately only two axial concentration profiles were presented so a detailed analysis is not possible. The great majority of the reported results for the interchange coefficient were higher than the Davidson and Harrison (1963) model predicted.

In addition there is only sufficient data to compute concentration values and not enough to crosscheck by means of a mass balance. This is a frequent problem in the field which is probably due to the difficulties in obtaining measurements. It should be noted that the bed model used gave results for bubble diameter which were very close to those injected into the bed. However the model did have an important influence in the emulsion flow rate as the wake was taken into account. This is because the bubbles, at high speed in a small bed, carry a large fraction of the particles of the emulsion phase, and this influences the speed of the gas in the emulsion. The interchange coefficient is proportional to the emulsion flow rate as seen in Appendix (II-3). For reasonable wake size the exchange coefficient remains high.

It is questionable whether gas was sampled within the emulsion concentration as it is difficult to determine if no bubbles passed by the probe. An error analysis on this topic is presented in Appendix (III-1), showing that the error for this is usually small. Also, as Latham and Potter (1970) indicate, if the fraction of the emulsion and bubble sampled
is the same at each height, the slope of the logarithmic concentration profile does not vary. This is also shown in Appendix (III-1).

It should be noted that many authors (Hovmand, 1969, Gueles de Cravahlo and Harrison, 1975, Toor and Calderbank, 1967) argue that the end effects predominate in these experiments. Walker (1975) counters this argument by claiming that diffusion must have been enhanced by the small particles.

The end effects may well have existed as they actually are a problem in this sort of experiment, as has already been indicated. However, the behavior within the bed, a number of centimeters above the distributor, should be as indicated by the equations. It is possible that the measurement technique may not have been accurate. On the other hand, Walker's (1975) comment has some merit. Consider a bubble travelling through a bed at more than 300 um/s. Then, for the kind of bubbles experimented with the cloud is very thin (about 2-5 particle diameters thick). In this situation the particle motion itself must be rather violent and mixing of the cloud and emulsion must be large. Consequently, not only should the exchange models based on a large cloud emulsion resistance break down, but very possibly the bubble models themselves. This has been noted by Grace (1971).

It should be remarked that even in the Davidson and Harrison model the diffusion term of the exchange coefficient predominated for these experiments. As will be seen, the more careful measurements of Pereira and Calderbank, while working with similar particles, have a very different behavior and thus possibly upset these arguments.
Data Reduction of a Set of Experiments of Davies and Richardson (1966)

Figure III-2
Reported Data Reduction of a Set of Experimental Data by Davies and Richardson (1966)

Figure III-3
Davies and Richardson (1967) later argued that the enhancement of the exchange must have been by cloud and wake shedding. This would be in accordance with contributions of Toei et al (1968) and Rowe et al (1969).

The exchange coefficients for the data is given in Figures (2) and (3). Note that for the PVC and catalyst particles of Figure (3), which were not analyzed the interchange coefficient seems to increase with bubble diameter. More recently, Rietema and Hoebink (1975) have presented work with similar results, also for very small particles (66 and 51μ) and low \( u_{mf} \)'s (0.20–0.22 cm/s).

It is difficult to decide upon the validity of these experiments. It is true that it is the most rudimentary of those to be analyzed and, consequently the one to be considered as the least reliable. However, as the available data is very limited, its consideration is justified.

III-5.2 Stephens, Sinclair, and Potter (1967). The experiments to be described here are only those of Stephens' in a 15.24 cm diameter fluidized bed. For Sinclair's 5.04 cm diameter bed no complete data is available.

The set up is similar to Davies and Richardson's (1966). Tracer gas is injected into the bed, which already is slightly above incipient fluidization. The injected gas, rich in mercury vapor (with molecular diffusion coefficient of 0.125 cm\(^2\)/s according to the authors) is done so in a continuous fashion. This has the effect of creating many small bubbles at the height of the distributor which later coalesce as they rise through the bed. The dimensions of the bubbles as a function of height is not given, nor is the rate of coalescence. The authors argue that that aspect is irrelevant as the measurements were made in the upper part of the
Data Reduction of a Set of Experiments of Stephens et al (1967)

Figure III-4
bed, where there was no coalescence. A 'bubble density' is given for each run from which it is possible, together with the actual superficial and incipient superficial velocities and the bed voidage, to define a bubble diameter which determines the fluid mechanical properties of the bed.

Radial concentration profiles were measured in the bed, the average being taken at each height to represent the emulsion concentration. It was noted that after a given height and when the particles were small, the profiles were flat. This was not the case for the larger particles where no tracer was found near the walls half way up the bed. It should be noted that bubble gas was sampled along with the particulate phase, thus making measurements prone to error as the amount of bubble gas sampled is unknown. Fortunately many of the runs had a low bubble to bed volume fraction and the error was possibly reduced.

The emulsion concentration profiles for the smallest particles show an erratic behavior. At the bottom-most 5-8 cms they are very low and then suddenly jump to rather high values. Figure (5) shows a typical example. This behavior suggests that most of the coalescence is done at the jump and the behavior seen by others, such as Szekely (1962) and Chavarie and Grace (1976), of very large exchange at the bottom is not seen here. It is quite possible that there is a stage where a lot of violent mixing prevails. In this aspect the experiments should be similar to those of a freely bubbling bed. Due to this, Guedes de Cravahlo and Harrison (1975) and Chavarie and Grace (1976) have suggested that end effects predominated.
Emulsion Concentration Profiles for a Run by Stephens et al (1967)

Figure III-5

$u_{mf} = 0.39 \text{ cm/s}$

$u_o - u_{mf} = 1.385 \text{ cm/s}$
The calculation of the interchange coefficients for the small particles gives the results shown in Figure 5. The discrepancies of the different models are made clear by indicating the resultant coefficients arrived at from the data by applying the model of Section (II-3.1) with a wake fraction model $f_w = 0.3$, $f_w = 0$ and also those calculated by the authors. Again it should be noted that the wake fraction has a large influence upon the emulsion phase flow rate which in turn affects the calculated experimental value of the exchange coefficient. As the magnitude of the interchange coefficient for the largest bubble size is unexpectedly low in the case of $f_w = 0.3$, it was assumed that the values with $f_w = 0$ better approximated the measured value. However, as a wake is expected, doubt should be cast on both experiment and model. As seen in this case, the results are probably best approached by the model of Davidson and Harrison (1963). If one considers that bubbles have coalesced and are very fast, these results are not surprising.

For the large particles the concentration profiles are of the expected shape, as shown in Figures (6, 7, 3). Recall, however, that the radial concentration profiles are not flat. Computation of bubble size and velocity indicate that the cloud fraction $f_c$ was large, as Drinkenburg and Rietema (1973) indicate. Consequently the equations (II-4.13), for the model assuming the cloud concentration different from the bubble of emulsion phase concentrations was also applied with the help of Hughes' (1977) fluid mechanical model. The exchange coefficients $\kappa_{dc}$ and $\kappa_{ce}$ are those of Kunii and Levenspiel and so thus it is called the modified Kunii and Levenspiel model by Fryer and Potter (1972). Then the resultant cloud and
Concentration Profile for Experiment of Stephens et al (1967)

Figure III-8
Emulsion concentration profiles are weighted, by the volume fractions within the bed, essentially as suggested by Fryer and Potter (1972, 1975). The resultant profiles indicate that this corrected Kunii and Levenspiel model (1969) is as representative as the simpler Davidson and Harrison (1963) model. This stands to reason as the interchange between bubble and cloud in the former model is identical to the total interchange between the bubble and emulsion in the latter. Hence, for this example it is difficult to discriminate between those two cases.

In the case of the largest particles, where $u_{mf} = 25$ cm/s, the calculated cloud diameter is larger than the bed diameter and there is — according to the Davidson and Harrison model — cloud overlapping. As Stewart and Davidson (1967) criteria for slugging is not met, the data has not been rejected. Again, as should be expected, the Davidson and Harrison exchange model is the most adequate, as a large fraction of the gas sampled is that of the cloud.

As the superficial velocity of the bed in these experiments was slightly above incipient fluidization, it is possible that the interchange was enhanced because of this. Drinckenburg and Rietema (1973) have observed this phenomenon.

The results seem to confirm that exchange is enhanced by disturbances within the bed. These disturbances result in bubble interactions which should — qualitatively speaking — be similar to those occurring in a freely bubbling bed. Rowe (1971) and Rowe et al (1971) give descriptions of some bubble perturbations.
III-5.3 Pereira and Calderbank (1975), also presented by Calderbank, Burgess and Pereira (1975). This is doubtlessly the most accurate work done to date in the analysis of exchange from single bubbles.

The authors periodically injected single bubbles of helium from the bottom of an air fluidized bed of 15.4 cm in diameter. The particles were electrically conductant. The molecular diffusivity for helium, as given by Kunii and Levenspiel (1963) and Rowe (1971), is 0.68 cm/s. The bubble velocity, size and shape were measured with the aid of a sophisticated capacitance probe consisting of five contacts. By measuring time delays between the different contacts they deduce the above parameters as well as whether the probe is on the centerline of the bubble. The data is immediately relayed to an online computer which discriminates between the bubbles passing the probe squarely or on a side. For the ones passing squarely concentration measurements are made throughout the bubble's vertical axis.

Six successful bubble encounters with the probe at one height were assumed sufficient so as to define an average bubble and emulsion concentration. The authors report that the shape of the bubbles did not vary with height, confirming Goldsmith et al's (1975) previous observations. Unfortunately they do not indicate whether volume varied with height, which would have been of interest (the writer believes that this is implied). They do say, however, that the measured bubble volume was reduced by 7 to 10% from the volume injected. This may be explained by noticing that if a wake is carried up by a bubble, some of the gas injected may travel in the cloud so as to compensate for the decrease of flow in the emulsion
Data Reduction of Set of Experiments of Pereira & Calderbank

Figure III-9
phase. Richardson and Davies (1963) have observed the opposite effects, but with the bed initially not bubbling and a superficial velocity which must have been higher than that of minimum fluidization.

Profiles of bubble and emulsion concentration for eight different bubble sizes were published. The exchange coefficients which are calculated definitely favor the Chiba and Kobayashi model for the smaller bubbles, and the Davidson and Harrison model for two largest bubbles. The latter coincides with work by Hovmand and Davidson (1968) and Guedes de Cravahlo and Harrison (1975), with slugs which quite possibly these were. For the smaller bubbles the results seem to indicate that a diffusion mechanism is more adequate. Other authors, not reviewed here, such as Chavarie and Grace (1975), Fryer and Potter (1976), Chiba and Kobayashi (1970) have found similar results.

This work should be given special weight due to the quantity of measured data it reports. As a result it is possible to calculate the exchange coefficient directly from the equations of the mass transfer model represented by equation (II-15) in a number of ways as shown in Table III-3. The discrepancies on the results are relatively small, and the average of them is usually within 10% of that reported. The tracer continuity equation (II-1) was also checked and giving result with standard deviations which varied between 0.7 and 3 percent from the measured mean concentration.

Admittedly, the probe may have interfered with the measurements. However, this is probably small due to its reduced dimensions and the high velocity at which the bubble approached it. In order to check the bed model
used for the previous experiments one set of data was submitted to the
standard scrutiny which consisted of using the bed model to predict
bubble diameter and velocity. It was found that because the bubble
velocity was 20% larger than that observed, the interchange coefficient
calculated became very high. In fact, the result was closest to Davidson
and Harrison's (1963) model. This the standard bubble velocity equation
(II-5) for a single bubble should be suspect for bubbles of the kind
studied here. They report that these specific bubbles were slower than what
a so-called normal bubble should be. Also, some scatter appears at large
heights for the modified logarithm concentration profile becomes very
sensitive as the argument nears zero.

The low results for the exchange probably implies that the probe
did not interfere too much with the bubble and surrounding flow for other-
wise results would have had to be more homogeneous. Other valuable data
that this method gives is the concentration of the immediate surroundings
of the bubble. It is possible to check again assumptions of the models
related to this region.

III-5.4 Latham and Potter (1970). These are the only experiments to be
analyzed which were made in a freely bubbling bed. The setup is quite
ingenious. The bed used has the same dimensions as that of Stephens et al
(1967), the tracer also being mercury vapor. The tracer is injected 45.7 cms
above the distributor and designed in such a way that it is evenly distributed
at that height. The superficial velocity is higher than the critical velocity
at which the emulsion phase gas velocity $u_e$ starts to descend in order to
compensate for the uprise of the particles in the wake of the bubbles. In this manner the tracer gas descends and is exchanged with the bubbles, producing an axial concentration within the two phases.

The wake fraction, $f_w$, was calculated for the case when $u_e = 0$, which was found by increasing the superficial velocity until tracer was detected below the injector. The result was very high ($f_w = 1.0$) about three times larger than those reported by Partridge and Rowe (1966) and Woollard and Potter (1963). With Hughes' model a value of $f_w = 1.165$ was found. Note that $f_w$ is the ratio of the wake to bubble void volumes. It is possible that this tracer detection was earlier than it should have been because of molecular diffusion in the emulsion phase and downward gas flows in the emulsion as those reported by Calderbank et al (1975). Absorption to the downflowing particles may be disregarded as particles were glass spheres. Another possibility is that, as the other experiments just quoted refer to beds with single injected bubbles in incipiently fluidized beds, the particle upflow is increased with the presence of swarms of bubbles. Nonetheless, by no means may the difference be accounted by experimental error.

Radial concentration gradients were not flat, with a maximum difference between centerline and walls of almost 50% of the concentration at the injection level. The profiles became flatter the larger the distance from the injector.

Continuous sampling took place within the bed, presumably within the particulate phase, at different heights.

Some of the data reported was in the slugging regime, according to the Stewart and Davidson (1963) criterion, and consequently neglected.
Concentration Profiles for a Given Experiment of Latham and Potter (1970)

Figure III-10
Application of

eq. (II-14) eq. (II-15)

Davidson and Harrison
Kunii and Levenspiel
Chiba and Kobayashi

\[ u_0 = 4.75 \text{ cm/s} \]
\[ f_w = 1.0 \]

Concentration Profile of Fig. 10 for a Different Wake Fraction

Figure III-11
Concentration Profile of Fig. 10 for a Different Wake Fraction

Figure III-12
Concentration Profile for a Given Experiment of Latham and Potter (1970)

Figure III-13
Concentration Profile of Fig. 13 for a Different Wake Fraction

Figure III-14
From the data on the bed properties and the superficial velocities, the superficial velocities were calculated for different wake fractions and models. As the bed was freely bubbling it was necessary to obtain the bubble characteristics from Hughes' model. Two of the mass transfer exchange models were used: (2) and (3). The former, which assumes one mass transfer resistance and includes bubble volume change, seemed more adequate than the other, which neglects volume change because volume variations were considerable. As will later be explained it is at present best to neglect the volume variations. The cloud thickness was usually less than 5% of the bubble diameter and consequently bubble and cloud were assumed well mixed. The wake was assumed to be at the concentration of the cloud. This, of course, could be questionable, but proved to be more adequate than assigning the wake the concentration of the emulsion.

In Figures (10-12) the theoretical profiles for the case of $u_{mf} = 4.37 \text{ cm/s}$ are compared with data for different wake fractions. As seen in Figure (10), for $f_w = 1.0$ the value given by the authors for the critical superficial velocity, no model is superior. As the wake fraction dependence on variations of flow rates has not been studied, it is difficult to predict which is the most adequate. As the wake was assumed to be at the same concentration as that of the bubble (following the authors), the driving force between bubble and emulsion is artificially increased, which would favor a lower interchange coefficient. Also, if the wake fraction were to increase the Davidson and Harrison model (1963) would be favored. Consequently, although the results are by no means conclusive, the latter exchange model seems most adequate.
The same trends are found when comparing the theoretical models with data for the higher superficial velocities, as shown in Figures (12, 13 and 14). The difference lies in that all the models seem to underestimate the exchange.

The underestimation by all the theoretical profiles is increased if the volume change of the bubble is also taken into account by using Model (2) of the mass transfer equations for data reduction, as shown in Figures (11-13) by the thin lines. In fact, some underpredict the exchange so much that the tracer concentration is seen to increase with increasing height, which is unacceptable. The reason for this is that although bubble growth has been allowed for the theoretical models disregard transfer due to coalescence or change of volume of the bubble by interaction with the emulsion. In Appendix (LV-3) a form of the interchange is suggested and also it is shown that the more standard mass transfer equation (II-15) possibly is more adequate.

The main value of this work is that it shows the complexity of the behavior of bubbles in a freely bubbling bed and that the exchanger is higher, compared to that of single bubbles. Chavarie and Grace (1976) strongly emphasize that there must be important dissimilarities between these two types of flows.

III-6 COLLECTION OF DATA FROM PAPERS FOR MINIMALLY FLUIDIZED BEDS

Another point of view was taken in the search for an appropriate interchange model. At minimum fluidization, the three principal diffusion models have two dimensionless parameters in common. These two parameters are combined in different ways, according to assumptions inherent in each
model. According to the three models, these two dimensionless parameters are interrelated in the following manner:

for the Davidson and Harrison model:

\[ \pi_2 = a + b \pi_1 \]

for the Imai and Levenspiel model:

\[ \pi_2' = 45 + 3.85 \pi_1 \]

\[ \pi_2'' = 3.71 \pi_1^{0.5} \]

and

\[ \frac{1}{\tilde{\pi}_2} = \frac{1}{\pi_2'} + \frac{1}{\pi_2''} \]

for the Chiba and Kobayashi model [17]:

\[ \pi_2 = 5.71 \pi_1 \]

where in all cases,

\[ \tilde{\pi}_1 = \frac{1}{u_{\text{mf}}} \left( \frac{d_3^{1/2}}{d_0^{1/2}} \right)^{1/2} \]

\[ \pi_2 = \frac{\kappa \beta d_3}{u_{\text{mf}}} \]

Note that, as usual, the wake fraction has been neglected from Chiba and Kobayashi's model. Also, \( u_2 = 0.71 \sqrt{gd_3} \) has been assumed. Then plotting the theoretical curves, \( \pi_{\text{mf}} \) was assumed to be 0.5. Available experimental data can be plotted as functions of these two parameters, and compared to the predicted results of the various models. All the plotted experimental data is reportedly at minimally fluidized conditions.
The graph for the three dimensional experiments is shown in
Figure (15). The behavior of the two dimensional data is very similar.
The sets of data is reported in Appendix (III-2). For the three dimensional
graph, data from Calderbank et al, Walker, Davies and Richardson (as plotted
by Stephens et al), Stephens et al, and Drinkenburg and Rietema are used.
For the two dimensional plots, data was collected from Drinkenburg and
Rietema, Walker, Rowe et al, Toei and Matsuno and Chavarie and Grace and
may be found in Appendix (III-2).

Under minimally fluidized conditions the dimensionless terms have
the following significance

\[ \Pi_1 = \frac{\text{mass transfer by molecular diffusion}}{\text{mass transfer by convection}} \]

\[ \Pi_2 = \frac{\text{total mass transfer from bubble to emulsion}}{\text{mass transfer by convection}} \]

It should be remarked that the mass transfer by convection was deduced to
be proportional to the minimum fluidization superficial velocity \( u_{mf} \) by the
Davidson and Harrison model.

There are a number of observations which should be noted:

a. In general, the data lies below the Davidson and Harrison
prediction. The exception occurs when very low \( u_{mf} \) appears. Also, this
is the case of Davies and Richardson data, which has already been questioned.
The other data is that of Drinkenburg and Rietema, was done with very small
particles (60 \( \mu \)). As the Davidson and Harrison model includes no transfer
resistance from the cloud to the rest of the emulsion phase, their model
is probably quite good when most of the bed is enveloped in bubble clouds.
**Figure III-15**

Collection of Mass Transfer Data
\[ u_{mf} \text{ (cm/s)} \]

<table>
<thead>
<tr>
<th>Value</th>
<th>Reference</th>
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</tr>
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</tr>
<tr>
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<td></td>
</tr>
<tr>
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</tr>
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<tr>
<td>0.80</td>
<td>Walker (1975)</td>
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Key to Figures III-15, IV-1 and IV-2

Figure III-15 (cont.)
b. In general, the Kunii and Levenspiel and Chiba and Kobayashi models seem to indicate a minimum, especially is most of Walker's data is neglected. This may be done considering that, as Walker indicates, many of his measurements were in the slugging regime. This does not, however, explain the low mass transfer rates. For larger particles (i.e. high $u_{mf}$) the data is best approximated by the Davidson and Harrison model.

c. The data points corresponding to one set of experiments (i.e. $u_{mf}$ and D are kept constant) usually fall in a line which has a value of $\pi_1$ which is almost constant. The reason for this becomes quite clear, as the only parameter that is varies in a given experiment is $d_p$ which is raised to the $-0.25$ power.

What is intriguing of the data is that for an individual experiment the trend of data points is usually almost perpendicular to the theoretical predictions. However, one would expect that individual experiments would at least indicate the direction anticipated by the models. Alternatively, the mass transfer may follow the more conservative models for small bubble diameter and the Davidson and Harrison model for larger diameters. Although most of the data does show this trend, the larger bubbles may really be slugs, where the latter model does hold. This brings up two questions: first, whether the data available is reliable; and second, whether the parameters of importance are really those which have been plotted.

Obviously not enough data is available as it was possible to collect only about 150 data points from the literature. Whether these points are reliable remains open to discussion as has been already noted. It is clear
that the parameters plotted are of importance as the data is scattered between the predicted limits. However, it seems also clear that if the experiments made up to now are at all reliable, there may be a third fundamental parameter. A similar graph is given for the two dimensional case. The same comments hold in this case. In Appendix (III-2) is a listing of all the data points, and also a derivation of some of the two dimensional models is found in Appendix III-3.

III-7 SUMMARY

The available experimental work has been examined, some cases in detail with the use of the equations of Chapter II. All the experimental data for minimally fluidized beds was plotted in a single graph. Some trends became apparent, notably that the Davidson and Harrison model usually is a good approximation for bubbles of diameters which were an important fraction of the bed diameter (> 2/3) and for large particles (> 500 μ) systems with high particle Reynolds numbers; and the Chiba and Kobayashi and Kunii and Levenspiel mass transfer models are conservative approximations for smaller particles (< 500 μ). For very small particles (< 80 μ), it is possible that the Davidson and Harrison model is adequate.
III-8 NOMENCLATURE FOR CHAPTER III AND ITS APPENDICES

A_b, A_e - average cross sections of bed corresponding to bubble and emulsion [L^2]

C - tracer concentration [-]

C_b, C_c, C_e - tracer concentration in bubble, cloud and emulsion [-]

C_ave, C_ave - volumetric and point average tracer concentration [-]

C_o - tracer concentration at injection point [-]

D - molecular diffusion coefficient [L^2/T]

D_e - effective dispersion coefficients [L^2/T]

d_b, d_B - effective and measured bubble diameter [L]

d_c - cloud diameter [L]

D_p - particle diameter [L]

F_b, F_c, F_e, F_T - bubble, cloud, emulsion and total flow rates [L^3/T]

F_o - tracer injection flow rate [L^3/T]

F_c, F_w - cloud and wake to bubble volume fraction [-]

g - gravitational acceleration [L/T^2]

h - height, vertical coordinate [L]

Δh - distance between measurement within a bed [L]

K_a - bubble to emulsion mass transfer coefficient [L^3/T]

K_be - bubble to emulsion mass transfer coefficient, per unit bubble volume [l/T]

N - number of bubbles per unit bed volume [1/L^3]

N_c - rate of tracer transferred across the cloud-emulsion interface [L^3/T]

Q_c - volume of tracer transferred from bubble to emulsion [L^3/T]
\( r \) - radial coordinate [L]
\( r_b, r_B \) - effective and measured bubble radius [L]
\( r_c \) - effective cloud radius [L]
\( Re_p \) - average particle Reynolds number in cloud = \( 3u_{mf}d_p/\nu \)
\( u_b, u_{br} \) - bubble velocity and velocity of bubble in a stagnant bed [L/T]

\( u_{mf}, u_f \) - superficial and interstitial velocities in a minimally fluidized bed [L/T]

\( u_e \) - interstitial velocity in the emulsion phase [L/T]

\( u_o \) - superficial velocity in a fluidized bed [L/T]

\( u_\theta \) - tangential fluid velocities in the neighborhood of a bubble [L/T]

\( V_b \) - bubble volume [L^3]

\( w \) - potential function [-]

\( z \) - axial coordinate [L]

Greek Symbols
\( \alpha \) - \( u_{br}/u_f \) [-]

\( \delta \) - bubble to bed volumetric ratio [-]

\( \varepsilon \) - error in measurement

\( \varepsilon_{mf} \) - bed voidage at minimum fluidization

\( \theta \) - polar angle [-]

\( \nu \) - kinematic viscosity [L^2/T]

\( \phi \) - particle shape [-]

\( \psi \) - stream function [L^2/T]
III-9 REFERENCES TO CHAPTER III AND ITS APPENDICES


REFERENCES TO CHAPTER III AND ITS APPENDICES (Cont.)


REFERENCES TO CHAPTER III AND ITS APPENDICES (Cont.)


CHAPTER IV

STUDY OF THE GAS MASS TRANSFER FROM FAST BUBBLES

TO THE EMULSION PHASE III

Explanation of the Observed Phenomena, Recommendations
and Conclusions

IV-1 INTRODUCTION. An attempt is made to explain why the bubble to emulsion mass transfer coefficient varies as experimentally determined. The effect of fluid inertia forces become important for \( \text{Re}_p > 5 \), which at higher \( \text{Re}_p \) produce a laminar wake behind the particle, resulting in larger cloud to emulsion mass exchange than predicted by the diffusion models. Interactions and perturbations between bubbles also increases the mass transfer. The most relevant forms are by cloud overlapping and wake shedding. When using the diffusion models, a question arises about the adequacy of the use of the molecular diffusivity coefficient instead of an effective diffusivity coefficient which is then proposed. Recommendations are made for future experimental work as well as for modelling and data correlation. A bubble to emulsion model is recommended for temporary use which applies concepts presented in this chapter and developed in Appendix IV-3. A final conclusion to the work presented in Chapters II and III is then given, where it is emphasized that much more work is required in order to really understand and quantitively evaluate the phenomena which occur.
IV-2 DISCUSSION OF THE EXPERIMENTS AND THEORY

From the preceding chapters, it is apparent that the results have not shown conclusive evidence upon which exchange model is the most adequate representation of the mass exchange mechanism. On the one hand, the experiments dependent on a fluid mechanical model for the determination of important parameters seem to predict a high mass exchange and, on the other hand, the only work where all important parameters were measured predicts a low value except for the largest bubbles which probably were slugs. Even the commonly accepted trend that the coefficient decreases with bubble size has been recently challenged by a seemingly careful experimental work presented by Rietema and Hoebink (1975).

Most of the work has been done with very fine particles (around 100μ) which probably are not representative of mass transfer mechanisms for larger particles even in the fast bubbling regime. Consequently, only a qualitative discussion will be made in an attempt to help clarify the different mechanisms involved.

IV-2.1 The Convection and Diffusion Related Models. For fast bubbling regimes, as long as cloud overlapping is not present, the Davidson and Harrison model should, in principal, be inadequate. It has been shown experimentally by Rowe et al (1964) among others, that the gas rushing through the roof of a bubble is swept down along the walls of the void by the particles and back into the bubble. From their photographs it is clear that, for their experimental conditions, a very large portion of the gas recirculates within the cloud and hence exchange between cloud and emulsion
may by no means be represented by the sort of throughflow suggested by this model. However, as Toor and Calderbank (1967) and Pyle (1972) point out, the convective term becomes unimportant in comparison with the diffusion term for small particles. This diffusive term is very similar to that of Kunii and Levenspiel and Chiba and Kobayashi. In fact, in the former one $u_{br}$, the bubble velocity with respect to a stagnant fluid, was chosen instead of $u_b$; the bed voidage $\varepsilon_{mf}$ does not appear as the resistance was assumed to occur at the bubble-cloud interface, on the bubble side, and the multiplicative constant is different primarily because of different assumptions on the bubble shape. In the limit of very small particles for a minimally fluidized bed, the Davidson and Harrison model is between $1.02/\varepsilon_{mf}^{1/2}$ and $1.02/\varepsilon_{mf}$ times larger than the Kunii and Levenspiel and Chiba and Kobayashi models, respectively. As $\varepsilon_{mf} \approx 0.5$, the coefficient is between 1.45 and 2.05 times larger. Consequently, the behavior of the models for small particles depends on a theoretical assumption on bubble shape and porosity. It should be indicated that the virtue of the Davidson and Harrison model resides in that for the other limit, when bed particles are large and bubbles are slow, the convective term predominates. Hence, for these cases, a knowledge of the convective term is important.

The diffusive mechanisms, as suggested in the literature, should also be suspect. All models assume a well defined interface between the phases in question, at least one of them being in the particulate phase. Molecular diffusion is a process which takes place at the molecular level.
However, all derived bubble models consider averaged properties over a volume where interstitial (or particle) dimensions are comparatively small. This is stated clearly in studies of Darcy's law (e.g. Batchelor (1967)) which was applied by Davidson and Harrison, stressed by Jackson (1971) for the discussion of his model and implied by Murray (1965) when deriving his equations. Consequently, the flow in the interstices is not represented in detail by the bubble models.

If the particles around the bubble were not in motion, the appropriate diffusion coefficient to be used would be those characteristic of a packed bed. For $Re_p = 1 - 5$ ($Re_p$ is measured with respect to relative velocity), which is typical for small particles in the cloud region (see Appendix IV-1), the packed bed longitudinal and radial diffusion coefficients are probably similar to the molecular one, according to Sherwood et al (1976). For larger $Re_p (>100\mu)$ the diffusion coefficients become independent of the molecular one and vary linearly with velocity.

It should be remarked that the motion of the particles should enhance the mixing of the homogeneous gases. For low $Re_p$, when particles do not have a fluid wake attached, the fluid is greatly influenced by the motion of neighboring particles as seen in the problem of a sphere in viscous Stokes flow (see White (1974), Batchelor (1967)). In fact, the particle should push much of the fluid in front and pull the fluid behind it. This clearly would bring some of the gas in the emulsion phase into the cloud for incoming particles and vice versa for emerging particles. Happel (1958) has published a theory which describes how the fluid could
be carried by these particles in a fluidized bed. For higher $R_{p}$, a laminar wake should be formed behind the particles, which would accompany it into and out of the cloud partially validating the assumption that the tracer does not recirculate in the cloud of Davidson and Harrison. Grace (1971) has indicated in this respect that for small particle systems, when the cloud thickness is of the order of the diameter of the particle, the exchange must be by mixing of gases at the interface. Walker (1975) possibly thinks on the same lines when he claims that diffusion is enhanced by small particles.

IV-2.2 Bubble Interaction and Perturbations. An effect which may be important is the disturbance created by other bubbles. A bubble displaces a certain amount of particulate phase which may have an influence on the gaseous mixing. This would be especially true in a violently freely bubbling bed. Rowe (1971) describes several perturbations he observed.

Interparticle forces in the small particle beds within the cloud may have significant effects on the gas exchange.

Bubble coalescence, as already indicated, should be an important factor. During the process, the volume of the final bubble has been observed to be different than the total volume of the parent bubbles by Grace and Venta (1973). Werther (1976) has indicated the importance that the passage of gas through the emulsion from one bubble to the other for mass exchange with the particulate phase when in the process of coalescence.

If the cloud fraction, $f_{c}$, is an important fraction of the bed itself, throughput of gas should be predominant.
As seen in Fig. III-15, the gas exchange coefficient per unit area \((K_{be_b})\) is more sensitive to bubble diameter variations than predicted by the models. It is possible that this is due to the mentioned perturbations or exchange at the level of the bubble's wake.

In the case of the experiments with single bubbles, only Pereira and Calderbank (1975) measured the exchange exclusively from well behaved bubbles and their surroundings. It may be assumed that there was a minimum of disturbances within the bed due to other bubbles as the interval between two disturbances was three seconds for the larger bubbles and 1.5 seconds for the smaller ones. In the case of the larger ones, only one bubble was within the bed at a time. Bubbles which did not go through the center of the bed were disregarded. Consequently, disturbances did not affect the interchange measurements. For these cases \(Re_p \approx 1\) in the cloud and it seems that the molecular diffusion coefficients are adequate as predicted for packed bed. Of course, whether the Chiba and Kobayashi numerical coefficient is generally adequate may not be answered at this stage.

For Davies and Richardson's (1966) experiments the particle size is smaller than that used by Pereira and Calderbank, so \(Re_p\) in the cloud is smaller and mass transfer should be hindered. However, the typical cloud thickness is about 2-6 particle diameters (compared to about 5-15 for Pereira and Calderbank's work), so possibly the effects described by Grace (1971) become important. Also, the frequency of the bubble injection is higher and, as the particulate phase concentration was recorded,
the effects of bubble disturbances have been included.

In Stephens et al. (1967) work, disturbances should be important as these include coalescence and other effects described in the last paragraph, especially in the small particle system. For the larger particles measurements of the exchange coefficient included those taken within the cloud which again would give high interchange coefficients. Exchange could be enhanced because of higher $Re_p$ in the cloud bringing in fluid inertia effects.

Fig. (1) is a graph of the data already given in Fig. III-15 but with axes representing

$$Sh = \frac{K_{bc} d_b^2}{D} \text{ vs } Pe = \frac{u_{br} d_b}{0.711 (g d_b)^{1/2}}$$

This is the most appropriate graph for the Chiba and Kobayashi theory, which may be written as

$$Sh = 8.04 \varepsilon_{mf} Pe^{1/2}$$

However, the least squares fit program using these same dimensionless forms gives a power dependence of 1.0 on the Peclet number.

The fit is not much more satisfying than that given in Fig. III-15 as the scatter is approximately the same (up to about a factor of ten). However the sets of data for the different experiments prove to go in a common direction with the correlations, in contrast to what was observed in Fig. III-15.

An attempt was made to correlate the data by a least square fit using the most relevant parameters found from the dimensional analysis
Plot of Sherwood vs. Peclet Numbers for the Bubble to Emulsion Exchange Data for Minimally Fluidized Beds

Figure IV-1
Plot of Stanton Versus Modified $\text{Re}_p$ for Minimally Fluidized Bed Mass Exchange Data

Figure IV-2
given in Appendix IV-2. The form chosen is a reduced form of Eqn.

\[ \frac{K_{be}d}{u_b} = H \left( \frac{D}{u_f d p} \right)^k \left( \frac{u_b}{u_f} \right)^\lambda \left( \frac{d_p}{d_b} \right)^m \left( \frac{u_f d p}{\nu} \right)^n \]

The fit gave a value of \( k = -0.15 \), which is physically unacceptable, as molecular diffusion cannot inhibit mass transfer. Consequently, the diffusion term was neglected and the fit which is plotted is given in Fig. (2), the calculated fit is approximated by

\[ \frac{K_{be}d}{u_b} = 4.72 \left( \frac{u_b}{u_f} \right) \frac{d_p}{d_b} \left( \frac{u_f d p}{\nu} \right)^{1/2} \]

with a scatter of about a factor of 6. It should be noted that all the terms on the left hand side originate in the Navier Stokes equation defined at the particle level. It must be concluded that there must be other important mechanisms of mass exchange besides molecular diffusion. The only experiments in which molecular diffusion was varied between runs in a bubbling bed are those of Drinkenburg and Rietema (1973). Four different species were used as tracers with diffusion coefficients varying in a factor of almost two. The other conditions in the bed were unchanged, but the average mass transfer coefficient data for the ethane tracer was consistently higher than for the methane tracer, although the latter has a higher molecular diffusion coefficient. The authors indicate that this is due to gas adsorption to particles. Also the scatter of the data was high. For slugs, Guedes de Crvalho and Harrison (1975) have found that
mass transfer increases when molecular diffusion increases. On the other hand, the experiments of Davies and Richardson (1966) and Pereira and Calderbank (1975) for comparable situations (minimum fluidization velocities and bubble diameters) but molecular diffusion coefficients five-fold different have similar mass exchange coefficients. This may be either due to experimental error, as explained before, or to the molecular diffusion coefficient being inapplicable.

Drinkenburg and Rietema (1973) have indicated earlier the inadequacy of the molecular diffusion coefficient. As there is no adequate means of calculating it, it is proposed in Appendix IV-3 to use the fixed bed diffusion coefficient instead.

In fact, Drinkenburg and Rietema (1973) have proposed on the basis of their measurements for $Re_p \approx 0.025$ effective radial coefficients two and a half times larger than molecular diffusion.

IV-3 RECOMMENDATIONS. Knowledge of the bubble to emulsion interchange coefficient is of great importance in the design of fluidized beds. Experimental work is very limited and the results of many are questionable. Theories which have been examined possibly show the right trends, but may be inadequate to explain mass transfer mechanisms satisfactorily. This is particularly true for a freely bubbling bed as the fluid dynamic behavior of interacting bubbles is virtually unknown. Some authors, like Chavarie and Grace (1976) and Chiba and Kobayashi (1970) believe that exchange may not be well represented by models considering single bubbles in an infinite space. However, it is futile to try to explain theoreti-
cally these mechanisms with so little data available. Both an experimental and theoretical approach must be taken. The following steps are recommended for all particle sizes and bubbling regimes of interest.

IV-3.1  Experimentation

1) An experimental study on the effective diffusion through a shear flow of particles is required in order to obtain a more appropriate knowledge of diffusion through the cloud. This could be done in a Couette type flow.

2) Undisturbed bubble exchange. Few experimentors have dealt with beds containing only one bubble. In this way the primary mechanisms of exchange could be elucidated, and would give a lower bound to the exchange coefficient in freely bubbling beds. The procedure suggested here is inspired by Pereira and Calderbank's (1975) work. Single bubbles should be injected into an incepiently fluidized bed. The frequency of injection would be so low that no tracer is in the emulsion phase when the bubble passes. A three dimensional bed would be required. A probe, with a computer on line, would be used to make all the required measurements, as in the case of these investigator's experiments. The detailed information given by the probe would eliminate the need of applying fluid mechanical models, which themselves have yet to be verified, as well as being a means of examining great numbers of modeling assumptions. The effect of bubble growth due to interstitial velocities higher than that of incepient fluidization, which has been reported by Drinkenburg and Rietema (1973) and Chavarie and Grace (1976) to be considerably higher,
may also be studied.

3) Bubble interaction. In the writer's knowledge it has not yet been understood how bubbles interact. Further search for papers should be done and only then some experimental work. This may best be done by using visual methods as those used by Rowe and coworkers in the early 1960's in a two dimensional bed. From the resultant flow patterns it will be possible to have a qualitative understanding of the mass transfer interactions between bubbles.

4) Interacting bubbles exchange. An identical setup as in (1), but in a freely bubbling bed. The method of injecting tracer within the bed without affecting the bubbles could be done either by back-mixing as Latham and Potter (1970) or by injecting it through a section of the distributor as Rowe and Evans (1974). Alternatively a chemical reactor could be considered. The probe already suggested in (1) would be used to measure all the relevant bubble properties. Pereira and Calderbank (1975) claim to be pursuing experiments with these characteristics and possibly many valuable suggestions may be obtained from their work.

IV-3.2 Modeling, Correlation and Collection of Data

1) Modeling. The theoretical bubble models having been exploited completely by the models analyzed and that given in Appendix IV-3, it will be necessary to consider the exchange at the level of the bubble wake, bubble irregularities, coalescence, splitting and more minor forms of interaction. It should account for the exchange coefficient per unit volume being less dependent than the predicted inverse proportionality on
bubble diameter. This may be related to variation of the stability of the bubble with size.

2) Correlation and collection of data. The more recent publications of data have proved to be the result of more careful work. It should be expected that this trend will continue so that future data will be more reliable and complete. Future data should measure these quantities: \( u_b, d_b, C_b, C_e \) and if possible, \( f_c, f_w \) and \( C_c \). Also data on \( d_p, u_{mf}, u_o \) and \( \phi \) should be reported. An empirical correlation with the use of dimensional criteria similar to that found in Appendix IV-2 could be attempted, as well as for verification and application of the mentioned models.

IV-3.3 Present Application of Models. In Table (IV-1) different regions of applicability of the models is suggested, depending on different properties.

When the bubbles and their clouds do not occupy a large fraction (about \( f_c \delta < 0.6 \)) of the bed, it may be assumed that the bubble interaction is not important and the models relying primarily on diffusion should be used. When the cloud is as thin as the concentration boundary layer originated at the cloud-emulsion interface, the Chiba and Kobayashi exchange model should be used. When the cloud is thicker than the concentration boundary layer the diffusion model developed in Appendix (IV-3) should then be used. The comparison of the cloud with concentration boundary layer thicknesses is done by comparing a pseudo Peclet number \( u_f d_b/D_e \) to the ratio of relative bubble velocity to interstitial velocity.
\( \alpha = \frac{u_{br}}{u_f} \). The effective diffusion coefficient should be taken to be 2.5 times the molecular diffusion coefficient when dealing with very small particles (about 100\( \mu \)) and low \( \text{Re}_p \) (<1) and for the other cases the one developed in Appendix IV-3. When the particle Reynolds number at the cloud-emulsion interface is larger than 24, mass transfer through the particle wake (as given in Appendix IV-3) should be accounted for. When the cloud to bed fraction is not negligible \( (f_c \delta > 0.1) \), the concentration within the cloud should be accounted for and may be taken as that of the bubble. It is suggested to assume it to be that of the bubble.

For larger cloud to bed fractions \( (f_c \delta > 0.6) \), bubble interactions should predominate, particularly in the form of cloud overlapping. In this case the Davidson and Harrison (1963) model is suggested as the concept of isolated clouds lacks meaning. Although not indicated in Table IV-1, the Davidson and Harrison (1963) model should also be applicable in the case where the cloud is less than 5 particles deep as the concept of cloud as a physical entity is tenuous.

Theoretically, for slow bubbles the throughflow term of the Davidson and Harrison bubble exchange model should be adequate, as seen in Chapter V. Data published by Cranfield and Geldart (1974) for slow bubbles which were used to calculate throughflow coefficients by Hughes (1977) confirm the Davidson and Harrison model.
**Suggested Mass Transfer Model**

**TABLE IV-1**

<table>
<thead>
<tr>
<th>Fast Bubbles</th>
<th>( \frac{u_{fb}}{De} &lt; \frac{u_{br}}{u_e} )</th>
<th>( \frac{u_{fb}}{De} &gt; \frac{u_{br}}{u_f} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>((1 + f_c)\delta &lt; 0.6)</td>
<td>(K_{be} = 6.77 \left( \frac{D_e}{d_b^3} \right)^{1/2} + K_1)</td>
<td>(K_{be} = 3.39 \left( \frac{D_e}{d_b^3} \right)^{1/2} + K_1)</td>
</tr>
</tbody>
</table>

| \((1 + f_c)\delta > 0.6\) | \(K_{be} = \frac{4.5}{d_b} \frac{u_{mf}}{u_{mf}} + 5.85 \left( \frac{D_e}{d_b^{5/2}} \right)^{1/2}\) |

| Slow Bubbles | \(K_{be} = \frac{4.5}{d_b} \frac{u_{mf}}{u_{mf}}\) |

\(K_1 = 0\) if \(Re_p < 24\)

\(K_1 = \frac{4.5}{d_b} \frac{u_{mf}}{u_{mf}} \frac{\alpha}{[(\alpha+2)(\alpha-1)^2]}^{1/3} \frac{1 - \epsilon_{mf}}{\epsilon_{mf}} \frac{0.40(\ln \frac{Re_p}{24})^2}{126 \ln \frac{Re_p}{24} + 1}\) if \(24 < Re_p < 130\)
IV-4 CONCLUSION. The subject of mass transfer between the bubble and emulsion phases has been studied. The methods used in the experimental work are open to many questions. Many times only a meager amount of data is taken which leads to use of doubtful assumptions (such as measurements taken in the emulsion phase only and the fluid dynamical behavior of the bed) that have a large influence on the calculations involved. Also, it is possible that the raw data reported is being misinterpreted. The theoretical contributions are still at a rather initial stage. All seem to more or less agree with the experimental data and it is hard to discriminate among them. This is compounded by difficulties in defining adequate boundary conditions.

If the experiments reported are at all reliable, for small particles the Chiba and Kobayashi (1970) and Kumii and Levenspiel (1969) models are conservative estimates. On the other hand the Davidson and Harrison model overestimates the exchange, and probably is the most successful for larger particles \( d_p > 500 \mu \).

Additional research is required in this area. It should be far more carefully done than was typical a few years ago, particularly in freely bubbling beds. In the modelling researchers should be aware that the mass transfer mechanisms may change for different bubble and particle sizes, even within the same bubbling regime. Due to the complexity of the process, theory should indicate the general mechanisms involved and the coefficients adjusted from experiment.

A new model for temporary use has been recommended: diffusion
type models when bubble interaction is unimportant and the Davidson and Harrison model when interaction is relevant. For large particle Reynolds numbers particles carry wakes which become an important form of mass transfer.

The practical importance of the mass transfer has not yet been entirely established for fluidized bed combustors as gas by-passing of the bed may be unimportant.

IV-5 SUMMARY. An explanation for the different physical bubble to emulsion mass transfer phenomena is proposed. A number of mechanisms are pointed out with which mass transfer should increase with respect to the Chiba and Kobayashi diffusion models. Unfortunately many are difficult to evaluate quantitatively. Particle wakes and bubble interaction (cloud overlapping wake shedding, etc) enhance the mass transfer exchange, explaining the success of the Davidson and Harrison model under some circumstances. The use of the molecular diffusion coefficient is suggested as inadequate to explain the diffusion process, in the same way as it is inadequate for systems such as packed beds. Recommendations for further work on the experimental and theoretical side are made. A mass exchange model is suggested for present use which covers most possible situations and based on the phenomena discussed in this chapter. It is concluded that far more work is required in this field.
NOMENCLATURE FOR CHAPTER IV AND ITS APPENDICES

\[ C_b, C_c, C_e \]  - tracer concentration in bubble, cloud and emulsion [-]
\[ D \]  - molecular diffusion coefficient \([L^2/T]\)
\[ D_{e}, D_{a}, D_{r} \]  - effective and effectual axial and radial dispersion coefficients \([L^2/T]\)
\[ d_b \]  - effective bubble diameter \([L]\)
\[ d_p \]  - particle diameter \([L]\)
\[ f_c, f_w \]  - cloud and wake to bubble volume fraction [-]
\[ g \]  - gravitational acceleration \([L/T^2]\)
\[ h \]  - vertical coordinate \([L]\)
\[ k_{be} \]  - bubble to emulsion mass transfer coefficient per unit bubble volume \([1/T]\)
\[ k_{be,d}, K_{be,w} \]  - bubble to emulsion mass transfer coefficient per unit bubble volume due to diffusion and particle wake \([1/T]\)
\[ Pe \]  - pseud Peclet number \(= \frac{u_{mf} d_b}{D}\) [-]
\[ Pe_a, Re_r \]  - effective axial and radial Peclet numbers \(= \frac{u_{f} d}{D_a}, \frac{u_{f} d}{D_r}\) [-]
\[ Q_p \]  - volume flow rate of particles through cloud \([L^3/T]\)
\[ r \]  - radial coordinate [-]
\[ r_b, r_c \]  - effective bubble and cloud radii \([L]\)
\[ Re_p \]  - average Reynolds number in cloud \(= \frac{u_{ave} d_p}{v}\) [-]
\[ Sc \]  - Schmidt number \(= \frac{v}{D}\) [-]
\[ Sh \]  - Sherwood number \(= k_{be} d_b/D\) [-]
\[ u_b, u_{br} \]  - bubble velocity and velocity of a bubble in a stagnant bed \([L/T]\)
\( u_{mf}, u_f \) - superficial and interstitial velocities in a minimally fluidized bed [L/T]

\( u_e \) - interstitial velocity in the emulsion phase [L/T]

\( u_o, u_r \) - tangential and radial fluid velocities in the neighborhood of a bubble [L/T]

\( u_{ave} \) - average interstitial velocity relative to particles in the cloud [L/T]

\( u_{ax} \) - velocity in the axial direction near the bubble [L/T]

\( V_b \) - bubble volume \([L^3]\)

\( W \) - potential function \([L^3/T]\) and particle wake fraction [-]

\( \Delta x \) - axial length incremental [L]

Greek Symbols

\( \alpha \) - \( u_{br}/u_f \) [-]

\( \delta \) - bubble to bed volume fraction [-]

\( \varepsilon_{mf} \) - bed voidage at minimum fluidization

\( \theta \) - polar angle [-]

\( \nu \) - kinematic viscosity \([L^2/T]\)

\( \phi \) - particle shape factor [-] on potential function \([L^2/T]\)

\( \phi' \) - modified potential function = \( r^2\sin^2\theta d\phi \) \([L^4/T]\)

\( \psi \) - stream function \([L^2/T]\)


CHAPTER V

RADIAL GAS DISPERSION BY SLOW BUBBLES

V.1 INTRODUCTION

Although the vast majority of experimental work on bubble-emulsion gas interchange has been done in the fast bubble regime, it is presently thought that fluidized bed combustors will work in the slow bubble regime, as indicated by Pyle et al. In fact, no literature has been found that presents either experimental results or theoretical models specifically on this topic. Consequently, a simple model has been created which is expected to at least indicate a lower limit for the gas dispersion within a slow bubbling fluidized bed. In a slow bubbling regime, for the most part the gas entering the bubble enters the emulsion phase, but does not recirculate in a cloud.

The gas interchange between the bubble and emulsion is largely a result of this convective through-flow, with possible influence due to a molecular diffusion mechanism, which is important in fast bubbling regimes. With this in mind, a simplified model that describes the dispersion of gas was considered. First it will be shown that the bubbles could be considered to be ideal mixing points, i.e., gas entering the bubble with an initial concentration difference will be well mixed when it leaves. In order to study the dispersion of the gas within a bed a simple model was then conceived consisting of stationary bubbles that supplied a short cut for the faster moving gas.
MODEL OF A SLOW BUBBLE AS A MIXING POINT

In order to see whether a bubble could behave as a good mixing point, it was modeled as a cylinder of radius \( r_o \) and height \( L \) through which a gas of axial uniform velocity \( v \) passed. The concentration profile \( C \) of the gas at the entrance was assumed known and only radially dependent (see Fig. 1).

The diffusion equation then reduces to that of Klinkenberg et al:

\[
\frac{D}{r} \frac{\partial}{\partial r} \left( r \frac{\partial C}{\partial r} \right) + D_a \frac{\partial^2 C}{\partial z^2} = v \frac{\partial C}{\partial z} \tag{v-1}
\]

but the boundary conditions are slightly different:

\[
C(r, 0) = C_o(r)
\]

A second boundary condition was taken, considering that the walls of the cylindrical bubble were something between (i) at constant concentration \( C(r_o, z) = C_w \) and (ii) adiabatic wall \( \frac{\partial C}{\partial r} \bigg|_{r_o} = 0 \).

\( D_r \) and \( D_a \) are the radial and axial diffusion coefficients which could be different if the flow was considered turbulent. It is shown in App. (v-1) that the diffusive term in the \( z \) direction, \( D \frac{\partial^2 C}{\partial z^2} \) is much smaller than the convective term in that direction, \( v \frac{\partial C}{\partial z} \), when \( \frac{vL}{D_a} \gg 1 \). It therefore may be neglected and so an extra boundary condition is unnecessary.

Depending on which boundary condition is used, the solution of the above equation is:
Fig. V-1 Cylindrical Bubble
\[ C(r,Z) = \sum_{i=0}^{\infty} \alpha_{i} J_{0}(j_{i}, i \frac{r}{r_{0}}) \exp \left[ -\frac{j_{i}^{2} D_{r}}{v r_{0}^{2}} z \right] \]

and

\[ (ii) \ C(r,Z) = \sum_{i=1}^{\infty} \alpha_{i} J_{0}(j_{i}, i \frac{r}{r_{0}}) \exp \left[ -\frac{j_{i}^{2} D_{r}}{v r_{0}^{2}} z \right] \]

respectively. The development of these solutions and the definition of the symbols are found in App. (y-1).

A measure of the mixing occurring in the bubble should consider the flattening of the emerging profile as compared to the entering one. This may be done by considering how each of the eigenfunctions of eqns. (2) independently decrease with \( z \). Then, as the first eigenfunction is the one that decreases least,

\[ \mu_{i} = 1 - \exp \left( -\frac{j_{i}^{2} D_{L}}{v r_{0}^{2}} \right) \]

may be considered a measure of mixing (\( \mu_{i} = 1 \) implies ideal mixing, and \( \mu_{i} = 0 \) implies no mixing). Figures 2 and 3 show the emerging concentration-profiles for entering profiles of the shape of these first eigenfunctions.

The values used were for the bubble and bed properties \( u_{mf} = 47 \text{ cm/s} \) and \( d_{b} = 7 \text{ cm} \) from a typical slow bubble of Cranfield and Geldart (1974). These were adapted by making \( v = 3 u_{mf} \), \( r_{0} = 3 \text{ cm} \) and \( L = 6.5 \text{ cm} \). The value for \( D_{e} \) was taken for CO\(_{2} \) in air from G. T. Davies' "Turbulent Phenomena" as \( D_{e} = D + D_{turb}^{\text{CO}_{2}} \), with \( D = 0.16 \text{ cm}^{2}/\text{s} \) and \( D_{turb}^{\text{CO}_{2}} = 19 \text{ cm}^{2}/\text{s} \). \( D_{turb}^{\text{CO}_{2}} \) is the eddy diffusion due to isotropic turbulence.
Fig. V-2

Exit Concentration profiles for constant wall concentration assumption

Fig. V-3

Exit Concentration profiles for impermeable wall assumption
It is seen that for the case of impermeable walls $\mu_1 = 0.79$ and for that of constant wall concentration $\mu_o = 0.46$. The concentration profiles for laminar flow are also given, although the flow most probably is turbulent as $Re = 3 u_{mf} d_b = 9000$, which is very much higher than the Reynolds number for turbulence in a pipe.

Davidson and Harrison (1963) show a figure of a slow bubble with a tracer gas entering it. The emerging gas appears to be well mixed. Reuter et al (1970) and Calderbank and coworkers (1975) have observed this in fast bubbles.

Figure (4a) is a plot of the mixing coefficient as a function of the modified Peclet number

$$Pe_M = \frac{2 v r_o}{D_e} \frac{2 r_o}{L}.$$  

Note that the factor $2r_o/L = 1$, and enters only because the bubble has been modeled as a cylinder. The ideal mixing point assumption does hold when $Pe_M \leq 80$, approximately.

As an example, the lowest mixing coefficient is predicted by the model for a bed of limestone as a function of bed diameter. The incipient fluidization velocity was evaluated by using Kunii and Levenspiel's (1969) eqn(3.20):

$$\frac{u_{mf} d_p}{v} = \left[ 33.7^2 + 0.0408 \frac{d^3_p (\rho_s - \rho_g) g}{\rho_g v^2} \right]^{1/2} - 33.7$$
Fig V-4a Mixing Coefficient As A Function of the Modified Peclet Number

Modified Peclet Number \( P_{em} = \frac{2ur_o}{De} \frac{2r_o}{L} \)
The lowest mixing coefficient for a given particle diameter is found for the slow bubble with the largest possible dimensions (for that particle diameter). This occurs when the bubble is at the transition point between slow and fast bubbling. The bubbles' diameter is thus found:

\[
\frac{u_{mf}}{\varepsilon_{mf}} = 0.711 \ (gd_b)^{1/2}
\]

Assuming, again, that the velocity of the fluid in the bubble is three times \( u_{mf} \), and that \( L = 2r_o = d_b \), the mixing coefficient becomes

\[
\mu_i = 1 - \exp\left( -\frac{4}{3}j_0.1 \left( \frac{D_r}{u_{mf} d_b} \right) \right)
\]

The properties used were taken as: \( \rho_g = 1.2 \times 10^{-3} \text{ g/cm}^3 \), \( \rho_s = 2.8 \text{ g/cm}^3 \), \( \nu = 0.165 \text{ cm}^2/\text{s} \), \( \varepsilon_{mf} = 0.45 \), \( g = 981 \text{ cm/s}^2 \), \( D_r = 19 \text{ cm}^2/\text{s} \). The result is shown in Fig(4b). It is observed that slow bubbles in beds of limestone must be good mixing points if \( d_p < 0.05 \text{ cm} \). This is not generally true for bubbles in larger particle systems.

\( \text{V-3 MASS TRANSFER INTERCHANGE COEFFICIENT} \)

For slow bubbles with velocities moderately lower than that of the neighboring emulsion, the recirculation of a cloud may be neglected. The mass transfer from the bubble interface to the neighboring emulsion phase should then be best described by the Davidson and Harrison coefficient as Pyle (1972) has
already indicated.

As is described in Chapter II, this model consists of assuming that exchange is due to a convective term added to a molecular diffusion term evaluated at the void-emulsion surface. Of the two terms, it may be said with confidence that the diffusion mechanism may be neglected. This may be confirmed by using the numbers of the previous paragraph from Cranfield and Geldart's paper, as well as a diffusion coefficient of helium \((D = 0.68 \text{cm}^2/\text{s})\), for example. Then, for the Davidson and Harrison model

\[
\frac{\text{Diffusion}}{\text{Convection}} = 1.3 \frac{D^{1/2} g^{1/2}}{u_{mf}^{1/4} d_b^{1/4}} = 0.078
\]

Consequently the diffusion mechanism may be neglected and the mass interchange from bubble to emulsion assumed to be exclusively by gas through-flow.

V-4 SLOW BUBBLE BED DISPERSION MODEL

As slow bubbles have a very high throughflow, and if bubble density within a fluidized bed is high, it is probable that most of the gas, going through the bed will use the bubbles as a short cut, as Kunii and Levenspiel (1969) have already suggested. Hence a model was conceived where the gas was assumed only to go through the stationary bubbles.

This requires the bubbles to be closely packed, as shown in Fig. 5 for otherwise the gas within the emulsion will bypass them. In
Figure v-5
Bubble Distribution and Gas Streamlines for $u_{br}/u_f \rightarrow 0$
that case the main mechanisms of dispersion within the bed
would probably be by particle mixing as well as by the diffusion mechanism
similar to those of packed beds. The bubbles will be assumed to be dis-
tributed throughout the bed in horizontal layers located on a square grid.
Every second horizontal layer is located such that their bubbles are on
the same vertical lines within these layers. However, the immediately
adjacent layers are displaced such that the bubbles appear at the center
of the square grids formed by the other layers (a "body-centered-rectan-
gular prism" distribution). This is best seen in Fig. 6. It is then
assumed that the gas going through a bubble must then go through either
the bubble vertically above it, two layers ahead or through one of the
four bubbles above it, in the adjacent layer and with an equal probability,
(refer to Fig. 7). Then it may be found what fraction of the gas at a
bubble at \((m_2, n_2, h_2)\) has come from bubble \((m_1, n_1, h_1)\) which is in some
lower horizontal layer, i.e. \(h_1 < h_2\). This fraction may be given as a
ratio of concentrations

\[
\frac{C(m_2, n_2, h_2)}{C(m_1, n_1, h_1)} = \prod_{m,n,h}^{h-n} \prod_{t=0}^{h-2t} \prod_{t=0}^{h-2t-p} \prod_{t=0}^{p-m} \prod_{t=0}^{h-2t-p} \prod_{t=0}^{h-2t-p}
\]

where \((m+n+h)\) is even and

\[
m = |m_2 - m_1|, \quad n = |n_2 - n_1|, \quad h = |h_2 - h_1|.
\]
**Figure V-6**

Bubble and Gas Distribution within a Bed Assumed by the Model
Figure V-7
Gas Distribution Between Bubbles at Different Heights
If \( m+n=h \) is odd, then

\[
\frac{C(m_2, n_2, h_2)}{C(m_1, n_1, h_1)} = (1 - 4 \xi) P_{m,n,h-1}
\]

The development of these formulae and that for the corresponding two-di-
dimensional bed is given in Appendix V-2.

The fraction of the gas going to one of the four bubbles above it may be found as a function of the distances between bubbles and of its diameter \( d_b \) if we assume a model for the flow around a bubble. If Davidson and Harrison’s model is assumed and the distance between hori-
izontal layers \( l_2 \geq 2d_b \), then the emerging flow from a bubble at layer \( l_2 \) (cms) below has expanded into a cylinder of constant diameter \( \sqrt{3} \ d_b \) as shown in Fig (5). In this case \( \xi \) will then be independent of \( l_2 \). The values for \( \xi \) have been calculated for these conditions and will be found in Appendix V-3. Fig. (8) is a plot of this function (\( \xi \) vs. \( l/d_b \)).

It is now possible to find the dispersion of gas from a "bubble-
source" if the dimensions of the grid and the bubble diameter are known. The "bubble source" is the only bubble in an infinite domain from which the tracer rich gas disperses through to the bubbles higher up. Fig. (9) is a graph showing the different concentration profiles at different bubble layer heights for a typical \( \xi \). Fig. (10) shows where the concentra-
tion is 1% of the axial concentration at that height for different values of \( \xi \). It may be seen that even for the case of maximum dispersion, \( \xi = 0.25 \) (i.e. no flow escaping through to the second layer), it is still
Figure V-8

Variation of the Probability Factor with Bed Horizontal Geometry When $\lambda_2/d_b \gg \sqrt{3}$
Figure V-9

Concentration Profiles at Different Bubble Heights
Figure V-10
"Boundary Layers" for Different Probability Factors $\xi$
quite limited, even 15 bubble layer heights above the source.

Next the variance of the concentration profile as a function of bubble layer heights was approximately calculated. This criteria for measurement of dispersion is standard, as indicated by Bischoff and Levenspiel, Levenspiel and Kunii and Levenspiel. Unfortunately the equation has one parameter \( l_2 \) which has to be adjusted in order to be used. In a similar situation for a fast bubbling bed, Hiraki et al (1968) used an equation which, translated in our terms, is

\[
\frac{\eta}{2} = \frac{\pi d_b^2}{(4\delta)}. \]

However the equation is not applicable as it defines the bubble void fraction in terms of areas and not volumes. So, as no information has been found on slow bubble dispersion a reasonable value must be chosen, such as \( l_2 = \eta l_1 \). The coefficient \( \eta \) should be larger than \( \sqrt{3/4} \) in order to satisfy the conditions of the model.

The variance, as given in Appendix V-4, may be equated to the variance of a continuous model (also found in Appendix V-4).

\[
\frac{D}{u} \frac{z}{u} = 2 \xi (64 \xi^3 - 16 \xi^2 + 4 \xi + 1) \left( \frac{\eta}{2} \right)^2 \left( h \right) \quad (V-3)
\]

where the left hand side refers to the variance of the continuous model and the right hand side to the variance of the proposed one. \( z \) is the height above the injection point and \( u \) is a characteristic velocity, taken here to be equal to \( (u_o - u_{mf}) \) in agreement with Rowe and Evans. Hiraki et al have assumed \( u = u_o \) and for Kunii and Levenspiel and Baerns et al it is \( u = u_o / \varepsilon_{mf} \). On the right hand side \( h \) is the number of bubble layers from the injection point which
corresponds to the height \( z \). So \( z = hl_2 \), and combining Eqn. (3) with Eqn. (AY-3-2)

\[
\frac{D_r}{u d_b} = \frac{48}{\pi} \xi (64 \xi^3 - 16\xi^2 + 4\xi + 1) \frac{l_1^4}{d_b^2} \delta.
\]

where \( 0 \leq \xi \leq 0.25 \) and \( \delta \frac{l_1^2}{d_b} \leq \frac{\pi}{24\sqrt{3}} \)

**V-5 COMPARISON WITH DISPERSION IN FAST BUBBLING FLUIDIZED BEDS**

In the knowledge of the writer, no information is available on dispersion in slow bubbling beds so it is necessary to compare with fast bubbling beds. Data of Rowe and Evans for 154 \( \mu \) particles and Schugerl's for 40,110 \( \mu \) and 250 \( \mu \) particles is considered. The bubble fraction \( \delta \), the bubble diameter \( d_b \) and the excess gas velocity \( u_o - u_{mf} \) are assumed the same in both cases. These properties were calculated from the fluid mechanical model presented in section II-3 and values given in the respective papers. For bubble frequency, Geldard and Cranfield's point frequency equation was averaged over the total height of the bed. The wake and cloud fractions \( f_w \) and \( f_c \), were neglected.

The ratio of the radial dispersion coefficient calculated here to the one found experimentally is plotted against the bubble fraction \( \delta \) in Fig. 11. The parameter \( l_2 \) was assumed to be twice \( l_1 \). For these calculations, \( 1.4 < l_2/d_b < 2.3 \). The assumption that \( l_2/d_b > \sqrt{3} \) made in the calculation of \( \xi \) is frequently violated, and so the ratio of radial dispersion coefficients is probably lower than that given.

Most of the data indicates that radial dispersion within a slow bubbling bed should be greater than in a fast bubbling bed. The disper-
Bubble fraction (δ)

Figure V-11

Comparison of Radial Dispersion Coefficient Fast Bubble Data With This Model
sion is increased significantly because the excess gas velocity is in-
creased as the density of the bubbles increases. Radial dispersion tends
to decrease in fast bubbling beds as particle size increases for the diff-
erent experiments.
If the dispersion in a slow bubbling bed is principally due to gas shortcircuiting, it will be at least comparable to the levels found in the fast bubbling beds. Of course, other mechanisms of dispersion have not been accounted for, such as the mixing of gases which does not take place within bubbles and especially those depending on adsorption of solids and the mixing of these. Furthermore, the throughflow is generally considered of a higher magnitude than that suggested by the Davidson and Harrison model, which was used. Higher throughflow coefficients would increase the dispersion as \( \xi (l/d_b) \) will be larger for a given \( (l/d_b) \) as the cylinders of influence are enlarged. However, the dispersive capability is reduced as the distance the gas travels from bubble layer to bubble layer is really larger than \( \lambda_2 \) due to their upward motion. Consequently, the concentration profile should be quite peaked for a considerable height above the injector. The model itself has only been developed to treat the case of a single source in an infinite domain as its purpose is to study a mechanism of dispersion within a slow bubbling fluidized bed. It may easily be extended to account for wall effects in the form of reflections and multisource situations.

The model is only valid when the concept of a bubble being a perfect mixing point remains reasonably valid i.e. when the modified Peclet number, \( Pe_M \) is less than 80. The assumption that gas through flow is \( 3 u_{mf} \) per unit bubble cross section is only valid when the bubble velocity \( u_b \) is significantly less than the gas velocity in the emulsion, i.e.
almost stationary bubbles. For this to occur, the bubble diameter must be small which is consistent with the assumption of small $Pe_m$.

The model for continuous dispersion may not be the most appropriate for the fast bubbling bed as the radial and axial dispersion coefficients are assumed equal. Hiraki et al. indicate that this is not applicable for $u_o > u_{mf}$. However, it was considered applicable to obtain a simple and rough estimate of the variance.

V -7 SUMMARY

A simple model for slow bubbling beds, of stationary bubbles distributed in a prismatic mesh or network was studied. The bubbles are assumed to be perfect mixing points and sole dispersive agents due to their property of shortcircuiting of gas. The model neglects effects such as mixing due to particle motion and is, therefore, conservative.

It may be concluded that this mechanism produces radial dispersion within the bed which is at least comparable in magnitude to that in a fast bubbling bed. For large particles, a slow bubbling bed may offer higher dispersion.
NOMENCLATURE FOR CHAPTER V AND ITS APPENDICES

- concentration [-]
\( C_o(r), C_w \) - concentration at base and wall of the cylindrical bubble [-]
\( C_i, C_e \) - axial inlet and exit concentration of the cylindrical bubble [-]
\( D \) - molecular diffusion coefficient [L^2/T]
\( D_a, D_r \) - axial and radial effective diffusion coefficients [L^2/T]
\( d_b \) - bubble diameter [-] [L]
\( f_c, f_w \) - ratio of cloud and wake to bubble volume [-]
\( h \) - number of layers of bubbles high from source in grid model [-]
\( J_k \) - \( k \)th order Bessel function [-]
\( j_{k,i} \) - \( i \)th root of \( J_k \) [-]
\( L \) - length of cylindrical bubble [L]
\( \ell_1, \ell_2 \) - smallest horizontal and vertical distance between bubble on grid [L]
\( m \) - number of bubbles in one of the two horizontal directions from the source bubble on grid [-]
\( n \) - number of bubbles in one of the two horizontal directions from the source bubble on grid, perpendicular to the \( m \)-direction [-]
\( Pe_M \) - modified Peclet number = \( \frac{4 V r_o^2}{D_e L} \) [-]
\( r \) - radial parameter [L]
\( r_o \) - radius of cylindrical bubble [L]
\( R(r) \) - radially dependent function from separation of variable of \( C(r,z) \) [-]
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>Re</td>
<td>Reynolds number inside bubble = ( v , d_b / u ) [-]</td>
</tr>
<tr>
<td>(v)</td>
<td>velocity inside cylindrical bubble [L/T]</td>
</tr>
<tr>
<td>Var</td>
<td>variance of slow bubbling bed model [L^2]</td>
</tr>
<tr>
<td>(u)</td>
<td>velocity used in the definition of the variance in the continuous model [L/T]</td>
</tr>
<tr>
<td>(u_b, u_{br})</td>
<td>absolute bubble velocity and bubble velocity in a minimally fluidized bed [L/T]</td>
</tr>
<tr>
<td>(u_{mf}, u_f)</td>
<td>superficial and interstitial velocities at minimum fluidization [L/T]</td>
</tr>
<tr>
<td>(u_o)</td>
<td>superficial velocity [L/T]</td>
</tr>
<tr>
<td>(z)</td>
<td>axial coordinate [L]</td>
</tr>
<tr>
<td>(Z(z))</td>
<td>axially dependent function from separation of variable of variable of (C(r,z)) [-]</td>
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**Greek Symbols**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>(\alpha)</td>
<td>(u_{br} / u_f) [-]</td>
</tr>
<tr>
<td>(\xi)</td>
<td>probability of gas element leaving bubble to enter a given neighboring bubble in the layer immediately superior [-]</td>
</tr>
<tr>
<td>(\mu)</td>
<td>gas mixing coefficient in cylindrical bubble [-]</td>
</tr>
<tr>
<td>(\nu)</td>
<td>kinematic viscosity of fluid [L^2/T]</td>
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CHAPTER VI

THE PROPER VALUES OF THE SHERWOOD AND NUSSELT NUMBERS FOR INDIVIDUAL PARTICLES IN FIXED AND FLUIDIZED BEDS

VI-1 Introduction

Since the pioneering work of Gamson et al [1] on evaporation of water from celite spheres into air more than three decades ago, a large number of investigators have studied mass and heat transfer from particles to surrounding fluids in fixed and fluidized beds. A number of different experimental techniques have been used for gases and liquids; a majority of the experiments were done under steady state conditions. Correlations and reviews of the experimental data are found in a number of reviews such as references [2] through [6]. For particle Reynolds numbers greater than 50 the trend of the particle Sherwood number and Nusselt number has been more-or-less well established. The Sherwood number is consistently higher than that given by the Ranz-Marshall equation for a single sphere in an infinite media [7]. This is also true for the Nusselt number.

At low particle Reynolds numbers experimental results have been highly controversial. Early experiments on packed beds indicated that the calculated Sherwood number approached zero as the Reynolds number became very small. Moreover, the results of various experimenters with gas varied by more than an order of magnitude. Recent experimental results, for example those by Hsiung and Thodos [8], have achieved higher values of Sherwood number at low Reynolds number. These results were achieved by more careful experimental procedures and by allowing for many of the possible sources of error, such as axial mixing in the flow. Nevertheless, recent data still indicates a decrease in the Sherwood number as the Reynolds number is
reduced toward zero. Mass transfer experiments between particles and liquids are inconclusive; for some the Sherwood number tends to zero at low Reynolds number while for other experiments the Sherwood number approaches a constant value. In general, most of the data for liquids yields a Sherwood number which tends to fall at small Reynolds numbers. However, for liquids the decrease in the Sherwood number commences at lower values of Reynolds numbers than for gases. Figure 1 shows a representative sample of experimental data which has been reported in the literature.

Heat transfer experiments at low particle Reynolds numbers have also tended to show the same kinds of behavior; the results for the heat transfer tests are further complicated because of conduction between adjacent particles.

Many investigators have attempted to explain the behavior of particle mass transfer in fixed and fluidized beds at low Reynolds numbers. Some authors have attempted to prove that the actual Sherwood and Nusselt numbers for an individual particle in a fixed or fluidized bed fall to zero as the Reynolds number decreases. On the other hand, other authors have attempted to explain the falloff of the Sherwood number by one of many mechanisms of experimental error. Other than the obvious fact that many investigators reached contradictory results the past work suffers from a number of shortcomings. Some authors neglect important mechanisms such as axial diffusion in the direction of fluid flow and inhomogeneities in particle packing across the cross section of the test section. Finally, a large segment of the published papers ignore the physics of fluids at low Reynolds number flow. For example, many authors attempt to use boundary layer concepts when the Reynolds number based on the particle
Fig. VI-1. Data and Correlations of Sherwood Numbers Derived from Experiments with Gases in Packed and Fluidized Beds.
diameter is less than or equal to unity. For very low Reynolds numbers, the boundary layer concept is totally invalid. As is shown in Schlichting [9] among others, when the Reynolds number is of the order of unity or less, derivatives in the flow direction are the same magnitude as derivatives lateral to the flow direction and all of the usual boundary layer approximations are inappropriate.

Recently a model formulated by Nelson and Galloway [10] has received considerable attention. This model is based on penetration theory using an approximation for the characteristic diffusion time, a well known concept at high Reynolds number. The Sherwood number predicted by this method again approaches zero for low values of Reynolds numbers. The Nelson and Galloway paper suffers from a number of major deficiencies. First, penetration theory is not appropriate at the low Reynolds numbers and, secondly the boundary conditions which Nelson and Galloway use are incorrect. The latter deficiency has also been pointed out by Schlünder [11]. The Nelson and Galloway results are difficult to accept because they do not even predict the correct trend of the Sherwood number with changes in voidage. Rowe [12] observed this and modified the result to properly allow for the variation with voidage. Its experimental confirmation was unsuccessfully attempted by Koloini et al [13]. In a later section Nelson and Galloway's work will be considered in detail.

Carberry [14] formulated a model which assumes a developing flat plate boundary layer is formed and destroyed at every particle diameter, \( d_p \). The solution showed that the Sherwood number approached zero for low Reynolds numbers. This solution is again invalid because the boundary
layer concept does not apply to these lower limits.

Zabrodsky [15] argued that the Sherwood number could not be negligible for low Reynolds number by formulating a model which had a thin shell of fluid surrounding the particle, the outer edge being at a constant concentration different than that of the particle surface. He then developed a relationship which showed a finite value of Sherwood number as the Reynolds number went to zero. This relationship has been corrected by Rowe [16] and also used by Gelperin and Einstein [17]. To explain the low Sherwood numbers found experimentally, Zabrodsky created a model where particles form an aggregation which permit local by-passing of fluids. The fluid does not bathe all the particles and the total heat transfer is reduced. This model of the fluid behavior is difficult to accept for packed beds and certainly will not apply in a fluidized bed system. Kunii and Suzuki [18] created a similar model where channeling was assumed to occur in preferred locations in a packed bed. Schlünder applied a model consisting of capillaries of two different diameters through which the fluid flows at different velocities. The mass transfer was modeled by using a solution analogous to the classical Graetz solution for a developing temperature profile in a pipe (also inappropriate for low Reynolds numbers). The models of Zabrodsky, Kunii and Schlünder neglected axial diffusion and arrived at low effective or experimental Sherwood numbers at low values of Reynolds number while retaining a finite Sherwood number for individual particles. However, in each case an unrealistic model of the bed geometry or the fluid flow behavior was employed to produce the low value of the effective Sherwood number. For example, in Schlünder's model with capillaries of dissimilar
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diameters, the ratio of the capillary diameters must be at least two or
three to produce any substantial decrease in the effective Sherwood number.

Finally Cornish [19] has further confused the issue by studying the
case of an infinite array of active particles in a stagnant fluid, taking
as the characteristic driving potential the potential difference between
the particle surface and the fluid at infinity. For a finite driving
force, the Sherwood number will naturally be zero.

Avedesian and Davidson [20] have formed a model at zero Reynolds
number for a single active particle in an infinite medium of inert particles.
The driving force is given by the concentration of the particle surface
and the fluid at infinity. This model should be more adequate for beds
with a small proportion of active particles.

Given the state of confusion that exists in understanding the
particle-to-fluid heat and mass transfer at low Reynolds numbers and
the growing concern in the topic in recent years, an attempt was made to
clarify some of the misconceptions in both the theoretical and experimental
work to date. The purpose of this chapter will be twofold; first to show
physically that at very low Reynolds number flows the Nusselt and Sherwood
numbers for individual particles should have values of the order of
magnitude of unity or above, and secondly, to construct a model to show
why experimental values of the Nusselt and Sherwood numbers are found to
be much less than unity. It will be demonstrated that without exception-
ally careful techniques, very low experimental values of the Sherwood
number will be derived from experiments at low Reynolds numbers even
when realistic system geometries are maintained.
VI-2 THEORY

VI-2.1 Proper Order of Magnitude of the Particle Sherwood and Nusselt Numbers

Consider the definition of the local heat transfer and mass transfer coefficients for the individual particles when all particles in the bed are active. This is defined as the heat or mass flux per unit area from the particle to the surrounding fluid divided by the characteristic driving potential difference. For heat transfer this driving potential difference is the surface temperature less the bulk temperature of the fluid in the flow channel adjacent to the surface. An analogous concentration difference is appropriate for mass transfer. The Nusselt and Sherwood number for an individual particle is then found from the product of the transfer coefficient per unit area of particle and the particle diameter divided by the fluid conductivity or diffusion coefficient respectively. At low Reynolds numbers the mechanism for the transfer of mass or heat laterally to the surface is diffusion. The greater the lateral distance on average that the heat or mass must be transferred from the surface to the fluid, the smaller the heat and mass transfer coefficient. Consequently, the lower limit for the transfer coefficients must be,

$$h > \frac{k}{\delta_{\text{max}}} \quad (VI-1)$$

$$k > \frac{D}{\delta_{\text{max}}} \quad (VI-2)$$

where $\delta_{\text{max}}$ is half the average interstitial distance between neighboring particles, see figure 2. Then,

$$Sh_p (Nu_p) > \frac{d}{p/\delta_{\text{max}}} \quad (VI-3)$$
Fig. VI-2. Characteristic Length for Mass and Heat Transfer Coefficients for Flow Through (a) Particles and (b) Tubes.
This expression is also true for laminar flow through ducts. For flow through pipes with a constant wall temperature, the well-known Graetz solution indicates that the Nusselt number approaches a constant finite value very far from the entrance length where the heat flux and the local surface to fluid temperature difference both go to zero. For this case, the correct value of \( \delta \) is 0.55 times the pipe radius. For other cross-sectional geometries, e.g., a square, triangle or rectangle, \( \delta \) is also approximately one-half the channel half width.

When the bed consists of a single active particle surrounded by inactive particles, the value of \( Sh_p \) and \( Nu_p \) for an active particle is somewhat altered. This case corresponds closely to a single particle in an infinite medium with allowances for tortuosity of the fluid between the inert particles and the void fraction, e.g. the Avedesian and Davidson model. Since for this case \( \delta \) can be larger than the interparticle spacing, the value of \( Sh_p \) and \( Nu_p \) given by the Avedesian model for a single active particle should be lower than the corresponding values for a bed with numerous active particles.

VI-2.2 Penetration Theory Results

Nelson and Galloway apply the penetration theory to a particle surrounded by a spherical shell of fluid of radius \( \frac{D}{2} + \delta \). Besides the typical penetration theory boundary conditions (constant surface concentration, constant initial fluid concentration), the authors assume that the outer boundary of the fluid shell may be assumed to be impermeable.
The equations are

\[ \frac{\partial C}{\partial t} = \frac{D}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial C}{\partial r} \right) \]  \hspace{1cm} (VI-4)

\[ C(r,0) = C_0 \]  \hspace{1cm} (VI-5a)

\[ C(\frac{d_p}{2}, t) = C_0' \]  \hspace{1cm} (VI-5b)

\[ \frac{\partial C}{\partial r} \bigg|_{\frac{d_p}{2} + \delta} = 0 \]  \hspace{1cm} (VI-5c)

They proceed to define the particle Sherwood number as

\[ Sh_p = \frac{\bar{N} d_p}{D(C_0 - C_0')} \]  \hspace{1cm} (VI-6)

where \( \bar{N} \) is the mass transfer rate per unit area of particle surface averaged over the period \( T \) that the fluid surface is renewed. In the paper \( T \) is taken to be

\[ T = 2.78 \frac{d_p^2}{v} \frac{Sc^{1/3}}{Re_p} \]  \hspace{1cm} (VI-7)

At low values of the Reynolds number, \( T \) increases, the fluid in the shell approaches saturation, and the mass transfer rate becomes small. The driving force used in the Nelson and Galloway definition of the Sherwood number is based on the initial concentration difference and it is invariant with time. Thus for low Reynolds numbers the Sherwood number based on equation 6 always goes to zero. In fact, as long as \( T \) is assumed to vary inversely with \( Re \), any particular form can be assumed
and Sh, defined by eqn. 6, will go to zero as Re decreases. It should be noticed that using a definition of the Nusselt number analogous to eqn. 6, i.e. the local heat flux divided by the initial bulk to surface temperature difference, the Graetz solution will yield a Nusselt which goes to zero at low Reynolds number and large entrance lengths! Thus the Sherwood number defined by Nelson and Galloway is erroneous.

To determine the correct mathematical limit of penetration theory at low Reynolds numbers, we have resolved the identical problem posed by Nelson and Galloway, eqns. 4 through 7, with a Sherwood number defined using the time averaged mass transfer rate and a time averaged driving potential, i.e. the difference between the surface concentration and the time averaged concentration at the outer shell of the fluid.

\[
Sh_p = \frac{\bar{N} d_p}{D\left(C_o' - C_2 + \delta_t\right)}
\]  

(VI-8)

A more detailed solution was required because the one presented by Nelson and Galloway does not directly give the time-varying concentration. Details of the new solution are given in Appendix VI-1. The results are shown on Fig. 3 for different values of the bed voidage. At low Reynolds numbers the Sherwood number, defined by eqn. 8, approaches a constant, finite value. The limiting value of Sh increases with decreasing voidage in agreement with experiments and contrary to the original results of Nelson and Galloway.
Fig. VI-3. Results from Penetration Theory where the Particle Sherwood Number is Defined by Eqn. (8).
Figure 3 illustrates that penetration theory cannot explain the observed behavior of Sherwood number with Reynolds number. However, as mentioned earlier, penetration theory is inappropriate at low Reynolds number; thus, the results given in Fig. 3 should not be regarded as an accurate prediction of the Sherwood number at low Reynolds numbers.

VI-2.3. Sources of Error

We are still left with the discrepancy between the true Sherwood and Nusselt numbers, which should approach values greater than or equal to unity at small Reynolds numbers, and the various experimental results which indicate that the Sh and Nu approach values one or more orders of magnitude below unity. When considering this discrepancy we must be careful to distinguish between the Sherwood number of an individual particle in a bed, Sh\textsubscript{p}, and the effective Sherwood number of a bed containing a large number of particles, Sh\textsubscript{e}. The former is defined as

\[
Sh\textsubscript{p} = \frac{N d_p}{D(C_s - C_B)} \quad (VI-9)
\]

where \( N \) is the instantaneous mass transfer rate per unit particle surface area, \( C_s \) is the instantaneous concentration at the particle surface and \( C_B \) is the bulk concentration of the fluid surrounding the particle. A time averaged Sherwood number can also be defined for an individual particle; for steady state it should be equal to the instantaneous value. The effective Sherwood number for a large bed can not be defined as easily. In its simplest form, consider a steady state mass transfer
process in a bed. If fluid flows through the bed in one dimensional plug flow, and there is no diffusion or backmixing in the flow direction, the concentration of the fluid leaving the bed, $C_e$, can be expressed as,

$$\frac{C_e - C_s}{C_i - C_s} = \exp \left[ - \frac{Sh_E A D}{d_p u A_{cs}} \right]$$  \hspace{1cm} (VI-10)

where $C_i$ is the concentration of the fluid entering the bed. $C_s$ and $A$ are the surface concentration and area of all the active particles in the bed, respectively, $u$ is the superficial velocity of the fluid, $A_{cs}$ is the bed frontal area, normal to the direction of flow.

Equation 10 can serve as a definition of the effective bed Sherwood number $Sh_E$. Most of the experimental measurements of $Sh$ are found by measuring the exit, entrance and surface concentrations for the bed and deriving the value of $Sh_E$. However, $Sh_E$, defined by eqn. 10 is only equal to $Sh_p$ when all of the assumptions needed for the derivation of equation 10 hold true; namely: steady one dimensional plug flow with no diffusion or backmixing in the flow direction. When these conditions do not hold, $Sh_E$ and $Sh_p$ will differ and the accuracy with which $Sh_p$ can be derived from experimental data depends on how closely the true physical conditions of the experiment are accounted for.

In the balance of this paper, we will examine low Reynolds number flow through a fixed bed which has a geometry typical of that used in experimental investigations. Conditions will be identified which tend to
cause large deviations between $\text{Sh}_E$ and $\text{Sh}_p$. Quantitative estimates will be made of the error in the derived values of $\text{Sh}_p$ when the actual physical conditions are not accounted for correctly or are omitted.

First, we will examine the consequences of non-uniform voidage or particle size across the bed surface. The voidages near the walls of a packed bed are considerably larger than the rest of the bed [21]. At low Reynolds numbers the fluid passing through the bed rapidly approaches the temperature and concentration of the particle surface, thus shallow beds must be employed to maintain a measurable difference between fluid outlet properties and particle surface properties. A shallow bed will tend to magnify any local anomalies in particle diameter or bed packing geometry. Figure 4 illustrates the results of a simplified model for the voidage variation across the bed cross-section for a constant pressure drop through the bed. The frontal area of the bed is divided into two regions each with a constant voidage in the flow direction. The ratio of the superficial velocities for the two regions was found by applying Ergun's equation to each region. The Reynolds number is based on conditions in the majority of the bed, which has a voidage of 0.4. Typically the region of the bed near the walls has a voidage of 0.7 or greater. At low Reynolds numbers, less than one-tenth, the superficial velocity in this region is twenty times* larger than the velocity through the core! When the bed diameter is one hundred times the particle diameter, the area of

*The resistance of the bed wall and radial mixing between regions of different voidage are not included, so this result should be taken as only an approximate value.
Fig. VI-4. The Velocity Ratio Between Two Regions of Different Voidages with the Same Applied Pressure Difference.
the bed within one particle diameter of the wall represents four percent of the cross-sectional area of the bed, but approximately forty-five percent of the total flow will pass through this area if the balance of the bed is assumed to have a uniform particle diameter and voidage.

Experimenters frequently manufacture their own particles which are not always identical; in a shallow bed particles of given sizes may possibly be found grouped together, creating regions of different flow rates in the bed. Particle size in certain regions may also be reduced by higher rates of sublimation. Figure 5 illustrates how the velocity varies between two regions which contain particles of different diameters. These results assume a uniform voidage across the entire bed. Note that a rather large diameter ratio must exist to achieve a large velocity ratio. It seems more likely that significant velocity ratios are caused by variations in the bed voidage rather than by differences in the particle diameter.

A second major source of errors may be due to the axial diffusion through the bed. At low Reynolds numbers, the axial diffusion is solely due to molecular diffusion. As the Reynolds number is reduced axial diffusion becomes progressively more important in comparison to convective energy or mass transport.

VI-3 PACKED BED MODEL

A model of a packed bed will be set up using a reasonable value of the individual particle Nusselt and Sherwood numbers, i.e. near unity at low Reynolds numbers. The model will be used to determine the value of
Fig. VI-5. The Velocity Ratio Between Two Regions of Different Particle Diameters with the Same Applied Pressure Difference.
Sh and Nu which would be derived from experimental results if some of the effects such as voidage variation and axial diffusion were improperly accounted for or ignored in the data reduction. If the effects are ignored the derived value of Sh and Nu corresponds to the effective value defined by equation 10. For these studies a bed with dimensions typically used in experimental investigations will be used in the model. The bed will be assumed to consist of two regions each with a constant fraction of the bed cross section, $a_1$ and $a_2$, for all heights, see Fig. 6. Each region has different but uniform voidage $\varepsilon_1$ and $\varepsilon_2$ and particle size $d_{p,1}$ and $d_{p,2}$ respectively. Fluid velocities through the two different regions are determined by assuming an equal pressure drop for each region for a fixed total flow rate. The velocities of the two regimes can be found by equating the pressure drop for the two regions using the Ergun relationship

$$
150 \frac{(1 - \varepsilon_1)^2 \mu u_1}{\varepsilon_1^3 d_{p,1}^2} + 1.75 \frac{1 - \varepsilon_1 \rho u_1^2}{\varepsilon_1^3 d_{p,1}^2} = \frac{(1 - \varepsilon_2)^2 \mu u_2}{\varepsilon_2^3 d_{p,2}^2} + 1.75 \frac{1 - \varepsilon_2 \rho u_2^2}{\varepsilon_2^3 d_{p,2}^2}
$$

and by conservation of mass of the incompressible fluid.

$$
\bar{u} = a_1 u_1 + a_2 u_2
$$

The particles are assumed to be spherical.

In the case of mass transfer, axial diffusion is accounted for by
CASE 1  \[ C_1(0) \neq C_2(0) \]  Eqn. 18
CASE 2  \[ C_1(0) = C_2(0) \]  Eqn B2

Fig. VI-6. Diagram of the Model of a Packed Bed.
using de Ligny's [22] effective Peclet number $\text{Pe}_j$ correlation for spherical particles. The Ranz-Marshall equation for mass transfer from a single particle will be taken to describe the particle Sherwood number in the bed. Only a fraction of the particles are assumed active. Diffusion between the two regions in the bed is neglected. Then, from reference 22, the Peclet number becomes,

$$\frac{1}{\text{Pe}_j} = \frac{\varepsilon_j}{\tau \text{Re}_{p,j} \text{Sc}} + \frac{0.7}{1 + \frac{5.8}{\text{Re}_{p,j} \text{Sc}}} \varepsilon_j \quad \text{for gases} \quad \text{(VI-13a)}$$

$$\frac{1}{\text{Pe}_j} = \frac{\varepsilon_j}{\tau \text{Re}_{p,j} \text{Sc}} + \frac{2.5}{1 + \frac{8.8}{\text{Re}_{p,j} \text{Sc}}} \varepsilon_j \quad \text{for liquids} \quad \text{(VI-13b)}$$

For heat transfer, axial diffusion will be defined by Votruba et al.'s [22] expression for an effective Peclet number

$$\frac{1}{\text{Pe}_j} = \frac{(k_{ez}/k_f)_j}{\text{Re}_{p,j} \text{Pr}} + \frac{14.5}{d_p (1 + \frac{5.1}{\text{Re}_{p,j} \text{Pr}})} \quad \text{(VI-13c)}$$

where, from Krupiczka [23],

$$\frac{k_{ez}}{k_f} = \frac{k_{s}}{k_f} \left[ 0.28 - 0.757 \log \varepsilon_j - 0.057 \log(k_s/k_f) \right] \quad \text{(VI-13d)}$$
\( d_p \) is given in \( \text{mm} \), and \( k_L \), \( k_s \), and \( k_e \) are fluid, particle and effective conductivities. From the Ranz-Marshall equation for a single particle, 

\[
S_{n_{p,j}} = 2 + 0.6 \left( \frac{D_{p,j}}{\epsilon_j} \right)^{1/2} \frac{Re}{\epsilon_j} \text{Sc}^{1/3} \quad (VI-14)
\]

The equation governing the mass transfer in the bed with axial diffusion is,

\[
\epsilon_{jD,e,j} \frac{d^2 C_j}{dz^2} - \frac{U_j}{\epsilon_j} \frac{dC_j}{dz} - 6(1-\epsilon_j) \chi \frac{k_j}{d_{p,j}} (C_j - C_s) = 0 \quad (VI-15)
\]

with boundary conditions: at \( z=0 \), the bed entrance,

\[
\left. \frac{dC_j}{d\varepsilon_{jD,e,j}} \right|_{z=0} = U_j C_j(0) \quad (VI-15)
\]

at \( z = L \), the bed exit,

\[
\left. \frac{dC_j}{dz} \right|_{z=L} = 0 \quad (VI-17)
\]

for \( j = 1, 2 \) which represent the different regions of the bed.

From the solution of these equations the mass average exit concentration \( C_e \) for the entire cross-section is then
\[
\frac{C_e}{C_s} = 1 - \frac{4d_p \Re_p}{\Re_p} \sum_{j=1}^{2} a_j \alpha_j \frac{\text{Re}_{p,j} \exp(\gamma_j)}{d_{p,j} [\alpha_j + 1]^2 - (\alpha_j - 1)^2 \exp(-\beta_j)}
\]  

(VI-18a)

with

\[
\alpha_j = \sqrt{1 + 24 \chi (1 - \varepsilon_j)} \frac{k_{D_e,j} \varepsilon_j}{d_{p,j} \varepsilon_j}
\]  

(VI-18b)

\[
\beta_j = \frac{u_j L}{\varepsilon_j D_{e,j}} \alpha_j
\]  

(VI-18c)

\[
\gamma_j = \frac{u_j L}{2 \varepsilon_j D_{e,j}} (1 - \alpha_j)
\]  

(VI-18d)

Possibly a more realistic case would be to require that the inlet concentration to the two regions be the same, i.e. \(C_1(0) = C_2(0)\). The solution is given in Appendix VI-2. For the range of values of the parameters used, it was found that the inclusion of this condition has an inconsequential effect on the derived Sherwood numbers. A third model is given in Appendix VI-3.

In the next section Sherwood numbers will be derived from the exit concentration and other data usually available to the experimenter. The core of the bed was assumed to occupy 95% of the bed cross-section \((a_1 = 0.95, a_2 = 0.05)\); the void fraction in the core, \(\varepsilon_1\), was taken as 0.4 which is typical for packed beds, and near the wall the void fraction
\( \varepsilon_2 \) was taken as 0.7. The bed was assumed to be five particle diameters high (\( L = 5 \, \bar{d}_p \)); all particles were assumed active (\( X = 1 \)); and the Schmidt number was taken to be approximately that of napthalene in air (\( Sc = 2.5 \)). The tortuosity \( \tau \), which appears in the equation for the Peclet number, will be taken as 1/0.7.

The choice of the values for the parameters was solely based on the criterion that the parameters have reasonable values, typical of those used in experiments reported in the literature, particularly those of fixed beds with gas as the fluid. No attempt was made to adjust the parameters to fit any specific experimental data since the purpose of the model is to account for the trends observed experimentally. For a given average particle Reynolds number, \( \bar{Re}_p \), the derived Sherwood number was found to be quite insensitive to variations of these parameters.

VI-4 **DERIVED VALUES OF SHERWOOD NUMBER**

The influence of channeling and backmixing was determined for each case considered by first calculating the "correct" exit concentrations from eqn. 18, using the Ranz-Marshall expression for the Sherwood number. "Experimental" values of \( Sh^* \) were found by using the correct value of \( C_e \) calculated from eqn. 18 and deriving \( Sh \) from a relationship containing the discrepancy to be investigated, see Fig. 7. For calculation of the experimentally derived \( Sh \) it will be assumed that the average Reynolds number \( \bar{Re}_p \), average void fraction \( \varepsilon \), and Schmidt number \( Sc \) are known.

*No subscript will be used for this quantity because in general it will not be either the correct individual particle Sherwood number nor the effective Sherwood number defined by equation 10.*
EXPERIMENTAL OPERATING PARAMETERS:

- Bed cross-sectional area and depth
- Particle diameter
- Particle concentration
- Voidage variation across x-section
- Gas thermophysical properties
- Inlet concentration & flow rate

CALCULATION OF "CORRECT" RESULTS

- Ranz-Marshall expression for Sh, including channeling & axial diffusion
- $C_e$, actual or experimentally observed exit concentration

CALCULATION OF DERIVED OR EXPERIMENTAL Sh

- Model with discrepancies e.g. neglect channeling, neglect axial diffusion, or experimental bias

EXPERIMENTAL Sh

Fig VI-7. Flow Chart for the Calculation of the Derived or Experimental Sherwood Number Used in this Paper to Examine the Influence of Various Discrepancies.
Logical definitions for these averages are:

\[
\overline{\text{Re}}_p = \frac{\overline{u} \overline{d}}{v} \tag{VI-19a}
\]

and

\[
\overline{\varepsilon} = a_1 \varepsilon_1 + a_2 \varepsilon_2 \tag{VI-19b},
\]

where:

\[
\overline{\varepsilon} = \left[ \frac{a_1 (1 - \overline{\varepsilon})}{d_{p,1}^{3/3}} + \frac{a_2 (1 - \overline{\varepsilon})}{d_{p,2}^{3/3}} \right]^{1/3} \tag{VI-19c}
\]

VI-4.1 Axial Diffusion

The first case to be investigated is the effect of ignoring the axial diffusion. To distinguish this effect from others we will assume the entire bed has a uniform voidage, \(\varepsilon_1 = \varepsilon_2\) in equation 18.

Figure 8 shows the experimental value of \(\text{Sh}\) when it is derived neglecting axial diffusion. In this case, the derived value is identical to the effective value given by eqn. 10. The solution with no axial diffusion is given by

\[
\text{Sh}_E = \overline{\text{Re}_p} \frac{\text{Sc}}{6(1 - \overline{\varepsilon})} \frac{\overline{d} \ln \frac{C_s}{C_e}}{L} \tag{VI-20}
\]

where the inlet concentration \(C_i\) is assumed negligible. Note that eqn. (10) and (20) are equivalent as \(A/A_{cs} = 6(1 - \overline{\varepsilon}) \chi L/d_p\).
Fig. VI-8. Derived Sherwood Number. Error: Axial Diffusion is Neglected.
Bed Conditions: \(a_1 = 0.95, a_2 = 0.05, L = \frac{5d_p}{\nu}, \epsilon_1 = \epsilon_2 = 0.4,\)
\(d_{p,1} = d_{p,2} = \bar{d}_p, \text{Sc} = 2.5, \frac{\chi}{\chi} = 1.0.\)
Fig. VI-9. Derived Sherwood Number. Error: Wrong Correlation for the Effective Axial Diffusion Used. Bed Conditions: $a_1 = 0.95$, $a_2 = 0.05$, $L = 5d_p$, $\epsilon_1 = \epsilon_2 = 0.4$, $d_{p,1} = d_{p,2} = \bar{d}_p$; $Sc = 2.5$, $\lambda = 1$. 
which is solved for \( \text{Sh} \) and \( \alpha \). \( \text{Pe} \) is given by eqn. (21) whereas \( \text{Ce} \) is found using equation (13a) and (18). The two expressions for \( \text{Pe} \), eqn. (21) and (13a) disagree at low values of \( \text{Re} \) where molecular diffusion predominates. At an \( \text{Re} \) of unity, \( \text{Pe} \) based on eqn. 13a is two times smaller than \( \text{Pe} \) based on eqn. 19 – at large \( \text{Re} \) they differ by a factor of four.

The derived \( \text{Sh}_p \) is shown on Fig. 9. In this case the error causes the derived value to be larger than the actual value by up to a factor of ten. Other correlations for \( \text{Pe} \) could cause the derived value to be lower than the actual one.

VI-4.3 Channeling – Voidage Variation

The previous results were obtained assuming the actual bed had uniform voidage throughout. In actuality, the voidage near the wall is higher than the voidage in the rest of the bed. This will be accounted for by assuming five percent of the bed cross-sectional area has a voidage of 0.7 while the remainder has a voidage of 0.4. The experimental value of \( \text{Sh} \) will be derived assuming that the entire bed voidage is at 0.4. Axial diffusion will be accounted for in the calculation of the actual value of \( \text{Ce} \) and in the derived value of \( \text{Sh} \) by using the same correlation for \( \text{Pe} \), eqn. 13a. Figure 10 shows how the derived \( \text{Sh} \) deviates from the actual value, the maximum discrepancy occurs at a Reynolds number of about one-tenth which is about the lowest value of \( \text{Re} \) which has been investigated for gases. The error due to channeling is moderated at still lower values of \( \text{Re} \) because axial diffusion begins to
Fig. VI-10. Derived Sherwood Number. Error: Channeling due to Voidage Variations. Bed Conditions: \( a_1 = 0.95, a_2 = 0.05, L = 5d_p, \)
\( \varepsilon_1 = 0.4, \varepsilon_2 = 0.7, d_{p,1} = d_p, d_{p,2} = 1.5d_p, Sc = 2.5, X = 1. \)
dominate over convection in the process of axial mass transport and both calculations use identical correlations for Pe. The effective value of Sh will be smaller than the derived value shown on fig. 10, since axial diffusion is accounted for in the derived value.

VI-4.4 Channeling - Particle Size Variation

As demonstrated earlier, variations in the particle diameter across the bed cross-section will cause deviations in the flow. Assuming the particle size varies by twenty percent from one region to the next, the derived Sh number again is reduced, see Fig. 11, but the effect is not as pronounced as voidage variations. The fall-off in Sh could be exaggerated by assuming larger variations in the particle diameter, but it is not reasonable to assume careful experiments would suffer from such an error.

VI-4.5 Measurement Errors

At low Reynolds numbers, the difference between the exit concentration of the fluid, $C_e$, and the saturation concentration of the fluid at the particle surface $C_s$ becomes very small, approaching in magnitude the standard deviation of the experimental measurement, $\sigma$.

If an experiment was repeated a large number of times, and the errors were random, one would obtain a Gaussian distribution of measured concentrations around the real exit concentration which would be the mean of the measured values. However, because of the complexity of experimental preparation many of the results were taken on experiments using a very modest number of samples. As the difference between $C_e$ and
Fig VI-11. Derived Sherwood Number. Error: Channeling Due to Particle Diameter Variations. Bed Conditions: $d_1 = 0.95$, $d_{2,1} = 0.05$, $L = 5d_p$, $e_1 = 0.4$, $d_{1,1} = 0.99d_p$, $p_2 = 1$, $Sc = 2.5$, $x = 1$.
$C_s$ approaches $\sigma$, some measurements of $C_e$ will exceed $C_s$. Investigators would be tempted to exclude these experimental results as erroneous. This would distort the random distribution and bias the mean of the measured exit concentration. The error between the biased mean exit concentration and the actual exit concentration would be,

$$C_{e,\text{meas}} - C_e = -\frac{1}{\sqrt{2\pi}} \exp\left[-\frac{1}{2} \left(\frac{1 - \frac{C_e}{C_s}}{\sigma}\right)^2\right] \frac{1 - \frac{C_e}{C_s}}{\text{erfc}\left[-\frac{1 - \frac{C_e}{C_s}}{\sqrt{2}\sigma}\right]} \quad (VI-23)$$

In addition, an error may also occur in the measurement of the saturation concentration $C_s$. Disregarding measurements of the saturation concentration below the mean exit concentration would lead to even higher errors. We will neglect this source of error; we will assume $C_s$ is measured accurately and only random errors in measuring $C_e$ occur.

Figure 12 shows the error due to biasing the mean exit concentration when the standard deviation in the measurements of the exit concentration is two percent of the saturation concentration. The derived $Sh$ depends strongly on the assumed parameters of the experiment which influence the degree of saturation of the gas leaving the bed. For example, when twenty percent of the particles are active ($\chi = 0.2$) and five rows of particles are in the bed, the derived value of $Sh$ starts to depart from the actual value at Reynolds number of about one. When all of the particles are active, much larger errors in $Sh$ occur, as shown in the figure.

When some of the above errors occur simultaneously the derived value of
Fig. VI.12. Derived Sherwood Number. Error: Exit concentration measurements inaccurate, with standard deviation $\delta = 0.02$ (2% of $C_e^2$). Bed Conditions: $u = \frac{1}{d_p}$, $L = 5d_p$, $e = e^3$, $X = 0.4$, $X = 0.72$. 

Average Particle Reynolds Number $\frac{\nu}{d_p}$. 

$Sc = 2.5$.
Sh is found to be between one to two orders of magnitude below the real one, as shown on Fig. 13. Also shown on Figure 13 are experimental values reported by various investigators. The large discrepancies between different investigators are probably due in large part to different measurement techniques, bed geometries, and methods of deriving Sh. As stated previously, we have not adjusted the parameters in an attempt to fit the results of any investigator. However, by using realistic values of the parameters, the trends of the results and the approximate magnitudes of our derived Sherwood numbers agree with the bulk of the experimental results.

For liquids in packed beds where the Schmidt number is much higher, the errors in Sh due to axial diffusion and channeling appear at much lower $\frac{Re}{p}$. This is seen in Fig. 14 where errors due to channeling and omission of axial diffusion are considered. All bed parameters are as usual except that the Schmidt number is increased to 1000. For liquids the errors in Sh appear at much lower values of the Reynolds number than they do for gases; this is in agreement with typical experimental results presented in the literature [30,31].

In heat transfer experiments an additional source of error is the particle-to-particle conduction. This can be accounted for by using an effective thermal conductivity which includes the effects of conductivity through the fluid as well as the particles as indicated in eqns. (13c) and (13d). Fig. 15 shows the derived values of the Nusselt number when several sources of error are present in comparison with experimental re-
Fig. VI-13. Comparison of Experimental Results and Correlations of Sherwood Number for Gases in Packed Beds with Values Derived from the Model with the Following Discrepancies: Neglecting Voidage Variations, Not Accounting for Axial Diffusion, and with Error in the Measurement of the Exit Concentration.
Fig. VII-14. Comparison of the Sherwood Number Derived from the Model for Gas and Liquid Systems. Errors: Channeling due to Voidage and Lateral Diffusion Left Unaccounted for. Bed Conditions: \( a_1 = 0.95 \), \( a_2 = 0.05 \), \( 1 = 5d \), \( e_1 = 0.4 \), \( e_2 = 0.7 \), \( d_1 = d_2 = d \), \( x = 1 \) and for liquids \( Sc = 1,000 \) and for gases, \( Sc = p_1^{2.5} \).
Fig. VI-5. Comparison of Experimental Results and Correlations of Nusselt Number for Gases and Liquids in Packed Beds with Values Derived from the Model with the Following Discrepancies: Neglecting Voidage Variations, Not Accounting for Axial Diffusion and with Error in the Measurement of the Exit Temperature. Bed Conditions: $a_1 = 0.95$, $a_2 = 0.05$, $L = 5d_p$ and $10d_p$, $\varepsilon_1 = 0.4$, $\varepsilon_2 = 0.7$, $\varepsilon_2 = \varepsilon_1 = d_2$, $\text{Pr} = 0.7$, $\chi = 1$, $K_s/K_f = 1000$ and $d_p = 1\ mm$ in equation $^{(13c)}$. 
results. Note that with a large effective conductivity, an inappropriate representation of axial conduction in deriving the Nusselt number leads to large errors. Typically, experimental results for the Nusselt number show a larger decrease at small Reynolds numbers than do results for the Sherwood number.

For fluidized beds, all the above errors should also be found in mass transfer and heat transfer experiments. In addition, the hydrodynamics of a fluidized bed is far more complex due to existence of inhomogeneties in the system. For example, bubbles can allow large gas bypassing causing severe channeling errors. Difficulties are also found in making measurements within the bed as it is frequently not well known whether particle or fluid properties are actually being measured. For this reason the experimental values of Sh have been found in general to be lower for fluidized beds than for fixed beds. By the same physical arguments used earlier for fixed beds, it can be concluded that the true value of individual particle Sherwood number and Nusselt number must also remain greater than or equal to unity for fluidized beds.

VI-5 - CONCLUSION

The experimental results for packed beds which indicate that Nusselt numbers and Sherwood numbers for individual particles approach zero at low Reynolds numbers are caused by improperly accounting for or omitting phenomena important at low Reynolds numbers. These include flow channeling due to high voidage along the walls of a test vessel, axial diffusion or backmixing, and inaccuracies in the concentration or temperature measure-
ments. Analyses such as Galloway and Nelson's penetration theory are inappropriate at small Reynolds number and, moreover, when used with a reasonable definition of Sh do not indicate a falloff of Sh to zero.

At low Reynolds numbers, the actual Nusselt and Sherwood number for individual particles in a fixed or fluidized bed approach a value equal to or greater than unity.

When the overall heat or mass transfer performance of a fixed or fluidized bed is to be determined at low Reynolds numbers, a careful consideration of channeling and axial diffusion must be included. The overall or effective Sherwood number and Nusselt number for the bed will be well below the values for individual particles.

The computer program used to give the above results as well as those for the model presented in Appendices VI-2 and 3 is found in Appendix VI-4.
NOMENCLATURE FOR CHAPTER VI AND IT APPENDICES

- Total surface area of active particles in the packed bed [m²]
- Cross sectional area of packed bed [m²]
- Fraction of cross section for region of the bed with respect to the total bed cross section
- Coefficients in eqn. AVI-1-2
- Solute concentration in the fluid [moles/m³]
- Bulk concentration of fluid in the neighborhood of a particle [moles/m³]
- Inlet and exit solute concentrations in the fluid for the packed bed model [moles/m³]
- Initial and particle surface concentrations in the Nelson and Galloway paper [moles/m³]
- Concentration of the solute in the fluid at saturation [moles/m³]
- Molecular diffusion coefficient [m²/s]
- Effective axial diffusion coefficient [m²/s]
- Particle diameter [m]
- Heat transfer coefficient [W/m²K]
- Mass transfer coefficient [m/s]
- Conductivities of fluid and particle [W/mK]
- Effective conductivity of the packed bed medium with no flow [W/mK]
- Height of the packed bed [m]
- Mass transfer per unit particle surface area [moles/m³s]
- Effective Peclet number = u dₚ / ε Dₑ
- Prandtl number = Cₚ μ/κ
- Radial variable [m]
- Particle Reynolds number = u dₚ / ν
- Schmidt number = ν/D
- Individual particle Sherwood number defined by eqn. (9)
Sh_e - effective particle Sherwood number in a packed bed defined by eqn. (10)

Sh - derived Sherwood number from the packed bed model

t - time variable [s]

t_d - time required for the penetration depth to be equal to the shell thickness [s]

T - surface renewal period [s]

u - superficial velocity [m]

z - vertical length variable

Greek Symbols

α - defined by eqn. (18b), (22b) or (AVI-3-2)

β - defined by eqn. (18c)

γ - defined by eqn. (18d)

δ - thickness of the fluid shell surrounding the particle [m]

δ_{max} - half of average interstitial distance between particles [m]

Δ - defined by eqn. (AVI-2-2g)

ε - void fraction of a region in the bed

ξ - defined by eqn. (AVI-2-2b)

η - defined by eqn. (AVI-2-2d)

κ - defined by eqn. (AVI-2-2c)

λ - defined by eqn. (AVI-2-2e)

μ - dynamic viscosity [kg/m s]

ν - kinematic viscosity [m^2/s]

ρ - penetration depth of the concentration boundary layer [m]
σ - standard deviation of measured concentrations
τ - tortuosity
Ψ - defined by eqn. (AVI-2-2f)
χ - fraction of active particles in the bed

Superscripts

- - average value throughout the bed

Subscripts

j - j'th region of the packed bed (= 1,2)
1,2 - regions 1 and 2 of the packed bed
VI-7 REFERENCES TO CHAPTER VI AND ITS APPENDICES


CHAPTER VII

THROUGHFLOW AND RECIRCULATION PROPERTIES OF ISOLATED AND SWARMS OF BUBBLES

VII-1 INTRODUCTION

As in most fluid mechanical problems the equation of mass conservation is fundamental in the modeling of a fluidized bed. Of particular importance for the two phase models is the flow of gas related to slow bubbles, i.e. those bubbles which are rising slower than the interstitial gas within the emulsion (α<1).

A fraction of gas in the bubble enters from and later exits into the emulsion without returning to the void, as shown in Figure 1. The total gas flow rate leaving the bubble permanently divided by the total gas flow rate through the emulsion at minimum fluidization across an equivalent bubble cross section is the throughflow coefficient m. Another fraction of the gas within the bubble flows into the particulate region and back into the bubble, also shown in Figure (l). This is analogous to the cloud region of a fast bubble. The space occupied by it is called the recirculation region. Its volume, per unit bubble volume, is the recirculation coefficient β. The gas in the recirculation region moves up in the bed at the bubble rise velocity.

A gas mass balance in a slow bubbling fluidized bed may be formulated in a simplified manner by combining both the throughflow and recirculation volume. These are calculated in this chapter.

The flow pattern of gas in a neighborhood of a bubble is influenced by other surrounding bubbles, particularly when they are close to one
Figure VIII: Sketch of slow bubble with recirculation region.
another, or their velocity approaches the interstitial gas velocity in 
the emulsion, because the recirculation regions become very large and tend 
to interact. An approximate analysis will be presented here to determine 
the effect of the interaction. The difficulties of solving for the flow 
pattern of a three dimensional swarm of bubbles appears to be insurmount-
able and so two dimensional horizontal array of cylindrical bubbles is 
studied instead (see Fig. 2). The influence of horizontally neighboring 
bubbles is made immediately evident in Fig. 2a. All the gas flowing 
through the bed must pass through the bubbles as the recirculation regions 
of neighboring bubbles are in contact, thus impeding rapid gas flow. The 
effect of bubbles above the array is assumed to occur only when they are 
a distance 2h apart as the recirculating regions then meet. In order to 
allow for three dimensional effects, correction factors are produced from 
the study of the case of the two and three dimensional isolated bubbles. 
A combination of the throughput and recirculation coefficients are used 
to define an effective throughput coefficient, which is then analyzed.

VIII-2 THROUGHFLOW AND RECIRCULATION PROPERTIES FOR THE SIMPLER CASES

Using the assumptions of some of the two phase models, analytical 
solutions may be found for the isolated bubble in two and three dimensions 
and the two dimensional horizontal array of bubbles. Their properties 
will be studied in this section.
Fig. (VII-2) Sketch of an Horizontal Array of Bubbles
VII-2-l  The Flow Patterns

Particularly due to their simplicity, the assumptions of the Davidson and Harrison model [1] will be maintained, i.e. the particulate region behaves as an incompressible inviscid fluid uninfluenced by the gas flow and the gas motion flows relative to the particles according to D'Arcy's law. The streamfunctions of the gas flow for the isolated two and three dimensional bubbles in a frame where the bubbles are stationary are:

\[
\frac{\psi}{u_f a} = (1-\alpha) \left[ 1 - \frac{1+\alpha}{1-\alpha} \frac{a}{r} \right] \frac{r}{a} \sin \theta \quad \text{(VII-1a)}
\]

and

\[
\frac{\psi}{2} = (1-\alpha) \left[ 1 + \frac{2+\alpha}{1-\alpha} \frac{a}{r} \right] \left( \frac{r}{a} \right) \frac{2 \sin^2 \theta}{2} \quad \text{(VII-1b)}
\]

respectively, where \( r, \theta \) are cylindrical coordinates with origin at the bubble centers of radii \( a \), as given in Fig. 1. Also \( \alpha = u_{br}/u_f \), where \( u_{br} \) is the velocity of the bubble and \( u_f \) of the emulsion for this case.

Collins [2] calculated the flow pattern around a two dimensional bubble rising midway between parallel walls. As he later indicated [3] the expression also represents the flow pattern around a horizontal array of cylindrical bubbles separated by the distance between the walls. The potential is given by

\[
\frac{\psi}{u_f a} = (1-\alpha) \frac{z}{a} - T \coth \frac{\pi z}{a}
\]
where

\[ T = \tan \frac{\pi a}{2b} + \alpha \tan \frac{\pi a}{2b} \]  \hspace{1cm} (VII-2)

\( b \) is the half distance between the bubbles, as shown in Fig. 2. Finally, \( z = re^{i\theta} \).

The streamfunction is given by

\[ \frac{\psi}{u_\tau a} = \text{Im} \left( \frac{\chi}{u_\tau a} \right) = (1-\alpha) \frac{y}{a} + \frac{T \sin(\pi y/b)}{\cos h\left( \frac{\pi x}{b} \right) + \cos \left( \frac{\pi y}{b} \right)} \]  \hspace{1cm} (VII-1c)

where \( x = r \cos \theta \) and \( y = r \sin \theta \). It should be noted that these streamlines do not produce exactly circular bubbles, but approximate them with less than a 2% error if \( a/b < 0.5 \) [2].

**VII-2-2 The Limiting Streamsurface**

The three properties to be calculated depend on the streamsurface which separates the fluid in the recirculation region from that in the emulsion. As the goal of this study is to analyze mass conservation when the bubbles are travelling at a slower speed than the interstitial gas, only when \( \alpha \) is between 0 and 1 will be considered. The cases of isolated two and three dimensional bubbles and of a row of two dimensional bubbles, the streamfunctions of which have already been given in eqns. (1), will be studied here.

The velocity at the \( \theta = \pi/2 \) surface in the particulate medium is perpendicular to it, i.e. the radial velocity \( u_r = 0 \). Within the recirculation region, at \( \theta = \pi/2 \), the angular velocity \( u_\theta \) is positive and in
the emulsion it is negative. Consequently the separating line on the 
θ = π/2 plane between the recirculation and emulsion regions is a stagnation 
point S, see Fig. 1. As \( u_\theta \) is proportional to \( \partial \psi / \partial r \), this must be zero at S. 
Hence \( \partial \psi / \partial r \bigg|_{\theta=\pi/2} = 0 \) will give the radial coordinate \( r_o \) of the point S. 
The position of the stagnation point, as expressed in cartesian coordinates, 
(\( x_o, y_o \)), for the two and three dimensional isolated bubbles is, respectively,

\[
\frac{y_o}{a} = \left( \frac{1+\alpha}{1-\alpha} \right)^{1/2}, \quad \text{2 dimensional bubble} \quad \text{(VII-3a)}
\]

and

\[
\frac{y_o}{a} = \left( \frac{1+\alpha/2}{1-\alpha} \right)^{1/3}, \quad \text{3 dimensional bubble} \quad \text{(VII-3b)}
\]

For both, \( x_o = 0 \).

For the bubble array there are two cases, depending on the value of 
\( \alpha \). If \( \alpha \) is less than a critical value, \( \alpha_c \), given by [2]:

\[
\alpha_c = \frac{1 - (\pi a/2b) \tan \h (\pi a/2b)}{1 + (\pi a/2b) \tan (\pi a/2b)} \quad \text{(VII-4)}
\]

then a stagnation point \( S \), in Fig. 2b, is again identified, as in the 
cases of isolated bubbles, by setting the derivative of the streamfunction 
with respect to \( r \) to zero at \( \theta = \pi/2 \) (or with respect to \( y \) at \( x = 0 \)).

Then

\[
\frac{y_o}{a} = \frac{1}{\pi a/b} \arccos(1 - \nu T), \quad \alpha < \alpha_c \quad \text{(VII-3c)}
\]
where
\[ v = \frac{\pi a/b}{1 - \alpha} \] (VII -5)

On the other hand, if \( \alpha > \alpha_c \), the recirculation region spreads across the emulsion, as in Fig. 2\( \alpha \), so all the gas flowing across the bubble layer must flow through the bubble. Then

\[ \gamma_o = b, \quad \alpha > \alpha_c \] (VII -3d)

The \( x \)-coordinate in this case will be defined in section 2.5.

Combining eqns. (1) and (3), the value of the limiting streamsurface \( \psi_o \) is readily defined. For the isolated two dimensional bubble:

\[ \frac{\psi_o}{u_f a} = 2(1 - \alpha) \frac{1}{2} \] (VII -6a)

for the isolated three dimensional bubble

\[ \frac{\psi_o}{u_f a} = \frac{3}{2} \left[ (1 - \alpha)(1 + \alpha/2) \right] \frac{1}{3} \] (VII -6b)

and for the array of two dimensional bubbles

\[ \frac{\psi_o}{u_f a} = \frac{1}{\nu} \arccos (1 - \nu T) + \left( \frac{2T}{\nu} - T^2 \right)^{1/2}, \alpha < \alpha_c \] (VII-6c)
and
\[
\frac{\psi_o}{u_f a} = (1-\alpha) b, \quad \alpha > \alpha_c
\]  \hspace{1cm} (VII-6d)

Eqn. (6d) was found by noting that as all the gas must now flow through the bubbles because \( \alpha > \alpha_c \); and so the streamfunction which limits the recirculation region must also be the streamsurface in the midplane between the bubbles at a large distance from the bubbles themselves (due to symmetry). This implies that
\[
\frac{\psi_o}{u_f a} = \lim_{x \to \infty} \psi(x,b) / (u_f a)
\]

VII-2-3. The Throughflow Coefficient

As indicated above, the throughflow coefficient \( m \) is defined as the total gas flow rate leaving the bubble permanently, divided by the total gas flow rate through the emulsion across an equivalent cross section.

The gas flow rate \( q \) across two streamsurfaces \( \psi_1 \) and \( \psi_2 \) may be found by noting that the flow across them is given by [1]:
\[
q = 2 \varepsilon_{mf} |\psi_1 - \psi_2|
\]
for a two dimensional case and
\[
q = 2 \pi \varepsilon_{mf} |\psi_1 - \psi_2|
\]
for an axisymmetric situation. The throughflow coefficient is then given by
\[
m = \frac{2 \varepsilon_{mf}}{u_{mf} a} \frac{\psi_o}{u_f a} = \frac{\psi_o}{u_f a}
\]
and
\[
m = \frac{2\pi \epsilon_{mf} \psi_0}{\pi a^2 u_{mf}} = \frac{2 \psi_0}{a^2 u_f}
\]
for the two and three dimensional cases, respectively. Note that use of the equality \( u_f = u_{mf}/\epsilon_{mf} \) has been made. Combining the above with eqn. (6), the throughflow coefficients are found and given in Table 1.

VII-2-4 The Recirculation Coefficient

As indicated above, the recirculation coefficient \( \beta \) is defined as the volume within the particulate region enclosed by the limiting stream-surface divided by the bubble volume. In order to calculate these volumes, it is necessary to solve explicitly eqn. (1) for one of the coordinates. Combining this with eqn (6) and using cartesian coordinates it is found that

\[
\frac{sx}{a} = (1 - \frac{y_s}{x})(\frac{ys/a}{1 - ys/a})^{1/2} \tag{VII-7a}
\]

where
\[
s = \left(\frac{1 - \alpha}{1 + \alpha}\right)^{1/2}
\]
for the two dimensional isolated case;

\[
\frac{x}{a} = \left(\frac{2 + \alpha}{1 - \alpha}\right)^{1/3} \left[\frac{(\phi)^{2/3}}{3 - \phi} - \frac{\phi}{2^{2/3}}\right]^{1/3} \tag{VII-7b}
\]
Table VII-1

Throughflow Coefficients

<table>
<thead>
<tr>
<th>Bubble case</th>
<th>Throughflow Coefficient m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isolated three dimensional</td>
<td>$3[(1+\alpha/2)^2(1-\alpha)]^{1/3}$</td>
</tr>
<tr>
<td>Isolated two dimensional</td>
<td>$2(1-\alpha^2)^{1/2}$</td>
</tr>
<tr>
<td>Horizontal array- two</td>
<td></td>
</tr>
<tr>
<td>dimensional</td>
<td>$\frac{1}{\nu} \arccos(1-\nu T) + \left(\frac{2T}{\nu} - T^2\right)^{1/2}, \alpha &lt; \alpha_c$</td>
</tr>
<tr>
<td></td>
<td>$(1-\alpha)b/a, \quad \alpha &gt; \alpha_c$</td>
</tr>
</tbody>
</table>

Table VII-2

Recirculation Coefficients

<table>
<thead>
<tr>
<th>Bubble case</th>
<th>Recirculation Coefficient $\xi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isolated three dimensional</td>
<td>$\frac{3}{2^{2/3}} \left(\frac{1+\alpha}{1-\alpha}\right)^{\frac{2}{3}} \int_0^{\alpha} \left[\frac{1-\phi}{2+\phi}\right]^{2/3} - \frac{1-\phi}{2^{2/3}}^{1/2} d\phi$</td>
</tr>
<tr>
<td>Isolated two dimensional</td>
<td>$[1-\arctan\left(\frac{\alpha}{\sqrt{1-\alpha^2}}\right) - \sqrt{1-\alpha^2}] \frac{1}{1-\alpha}$</td>
</tr>
<tr>
<td>Horizontal array- two</td>
<td>$\frac{1}{2} \left[ \frac{1}{\pi a} \int_{y_0}^y \ln(d+y^2-1) dy + \frac{y_c}{a} \sqrt{1-\frac{y_c^2}{a^2}} \right.$</td>
</tr>
<tr>
<td>dimensional</td>
<td>$\left. - 2\arctan\left(\frac{\sqrt{a^2-y^2}}{y_c}\right)\right]$</td>
</tr>
</tbody>
</table>
where

$$\phi = \left(\frac{1 - \alpha}{1 - \alpha/2}\right)^{2/3} \frac{y^2}{a}$$

for three dimensional isolated case; and for the two dimensional array:

$$\frac{x}{a} = \frac{1}{\pi a/b} \ln(d + \sqrt{d^2 - 1})$$

(VII-7c)

where

$$d = \frac{T \sin \pi y/b}{\psi - \psi_o} - \frac{(1 - \alpha) y}{u_x a} + \cos \pi y$$

(VII-8)

and $\psi / u_x a$ is given by eqn (5c) or (5d), depending on the value of $\alpha$.

The recirculation volume is, using Fig. 1

$$V_{SBHC} = V_{SBAC} - V_{BACH}$$

Then, for the two dimensional cases

$$\beta = \frac{2}{\pi a} \left[ \int_{y_c}^{y_o} x \, dy - \int_{y_c}^{y_o} \frac{\sqrt{a^2 - y^2}}{y} \, dy \right]$$

(VII-9a)

and for the three-dimensional case

$$\beta = \frac{3}{2\pi a} \left[ \int_{y_c}^{y_o} 2\pi y \, x \, dy - \int_{y_c}^{y_o} 2\pi y \, \sqrt{a^2 - y^2} \, dy \right]$$

(VII-9b)

where $y_c$ corresponds to the intersection of the limiting streamline with the bubble interface C in Fig. 1 and is given by
\[
\frac{y_c}{a} = \sqrt{1 - \alpha^2}
\]  
(VII-10a)

for the two dimensional isolated bubble;

\[
\frac{y_c}{a} = (1 + \alpha/2)^{1/3} (1 - \alpha)^{1/6}
\]  
(VII-10b)

for the three dimensional isolated bubble. See Fig. 1 for a graphical definition of \(y_o\) and \(y_c\). No explicit solution is available for \(y_c\) in the case of the array of cylindrical bubbles, but the solution is the root of eqn (7c) when \(y = y_c\) and \(x = \sqrt{a^2 - y_c^2}\). Combination of eqns (7-10) for the different cases yields the expressions for the recirculation coefficient \(\beta\) found in Table (2).

VII-2-5 The Normalized Height of the Recirculation Region

The height \(h\) of the recirculation region, shown in Figs. 2 and 3, divided by the bubble radius \(a\) is the normalized height \(\chi\). So

\[
\chi = \text{maximum } (x/a), \quad y_c \leq y \leq y_o
\]

where \(x/a\) is defined in eqn.(7). The analytical difficulties involved reside in that the maximum may not necessarily have zero slope, but may be found at one of the extremes. For the isolated bubbles the maximum is either at \(y = y_c\) or in between. The different possible situation for the horizontal array of bubbles is illustrated in Figs. 2 and 3. The normalized height \(\chi = h/a\) is then given by differentiating eqns (6) and setting them to zero. When the extreme is not on the midplane
Fig. (VII-3) Streamfunctions for Different Conditions of an Horizontal Array of Two Dimensional Bubbles
between bubbles for the horizontal array, the mentioned equation could not be solved explicitly. The easiest method is then to find the maximum of \( d(y) \), which is a solution to the equation:

\[
T \left[ \frac{\psi}{u_f} \right] - (1 - \alpha) \frac{Y}{a} \cos \frac{\psi}{b} + \left\{ \frac{T}{\nu} - \left[ \frac{\psi}{u_f} - (1 - \alpha) \frac{Y}{a} \right]^2 \right\} \sin \frac{\psi}{b} = 0 \quad (VII-11a)
\]

for \( y_c \leq y \leq y_o \)

or otherwise the maximum lies at \( y = y_c \) if

\[
y < 3, \quad \text{where} \quad \gamma = \nu T \quad (VII-11b)
\]

If \( \gamma \geq 3 \), then the maximum is at the midpoint between the bubbles:

\( y = y_o = b \). The final solution for all the cases is given in Table 3.

VII-2-6 **Comparison of The Properties of Two and Three Dimensional Isolated Bubbles**

It is convenient to study the ratio of the throughput and recirculation coefficients and the normalized height which are being analyzed for the isolated cases. In this manner it will be possible to analyze the influence of the change in the "dimensionality" of the problem on the properties being studied. Advantage will be taken of the insight obtained later when three dimensional swarms of bubbles are considered. These are shown in Figs. 4-6.

For most of the range of \( \alpha (<0.8) \), the ratio of the throughput coefficient decreases slowly, but eventually dips down to zero as it approaches 1 because the ratio is proportional to \( (1 - \alpha)^{1/6} \). This suggests
Table VII-3

Normalized Recirculation Coefficients

<table>
<thead>
<tr>
<th>Bubble case</th>
<th>Condition</th>
<th>Normalized height $\chi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isolated three dimensional</td>
<td>$\alpha \leq 0.820829934$</td>
<td>$[1-(1-\alpha)^{1/3}(1+\alpha/2)^{2/3}]^{1/2}$</td>
</tr>
<tr>
<td></td>
<td>$\alpha &gt; 0.820829934$</td>
<td>$0.271171676(\frac{1+\alpha/2}{1-\alpha})^{1/3}$</td>
</tr>
<tr>
<td>Isolated two dimensional</td>
<td>$\alpha \leq 0.618033989$</td>
<td>$\alpha$</td>
</tr>
<tr>
<td></td>
<td>$\alpha &gt; 0.618033989$</td>
<td>$0.300283106(\frac{1+\alpha}{1-\alpha})^{1/2}$</td>
</tr>
<tr>
<td>Horizontal array- two</td>
<td>$\gamma &lt; 3$</td>
<td>$\max\left[\frac{x}{a}(y_1), \frac{x}{a}(y_c)\right]$ where $x/a$ is defined by eqn(7c) and $y=y_1$ is solution of eqn(11a)</td>
</tr>
<tr>
<td>dimensional</td>
<td></td>
<td>$\frac{1}{\pi a/b} \ln(y-1+\sqrt{y^2-2y})$</td>
</tr>
<tr>
<td></td>
<td>$\gamma \geq 3$</td>
<td></td>
</tr>
</tbody>
</table>

Note: $d$ is defined by eqn(8)  
$\gamma$ is defined by eqn(5)
FIGURE VII-4 - Ratio of Throughflow Coefficients for Two and Three Dimensional Isolated Bubbles
FIGURE VII-5 - Ratio of Recirculation Coefficients for Two and
Three Dimensional Isolated Slow Bubbles
FIGURE VII-6 - Ratio of Normalized Recirculation Heights for Two and Three Dimensional Isolated Flow Bubbles
that two dimensional throughflow is reduced more rapidly than three
dimensional throughflow.

The variation of the recirculation coefficient and normalized height
ratios is also moderate for most or all the range of $\alpha$. Although the
numerators and denominators of the two expressions cover the whole
positive spectrum, their ratios remain reasonably uniform (except for that
of the normalized height as $\alpha$ approaches 1). This indeed implies that the
behavior of the flow patterns are generally similar for two and three
dimensional cases.

As $\alpha \rightarrow 1$ the normalized height of the two dimensional bubble increases
at a more rapid pace than for the three dimensional case. This should be
expected: in order for the ratio of the recirculation coefficient to be
finite as a three dimensional recirculation region expands in three
dimensions and the two dimensional one in only two. In fact in both cases
$\beta \sim 1/(1-\alpha)$ in the neighborhood of $\alpha = 1$, while $\chi \sim 1/(1-\alpha)^{1/2}$ in the two
dimensional situation and $\chi \sim 1/(1-\alpha)^{1/3}$ for the three dimensional one at
the same limit.
VII-3 THE THREE DIMENSIONAL SWARM OF BUBBLES

The analytical solution of the Davidson and Harrison potential equations for the flow pattern around a swarm of three dimensional bubbles appears to be very difficult to achieve. However, considering that the changes on the flow patterns of neighboring bubbles from the isolated case in both two and three dimensions are similar, an estimate of the flow properties of the three dimensional swarm may be inferred from those of a two dimensional swarm, i.e.

\[
\frac{\eta_{(\text{swarm, 3 dimensional})}}{\eta_{(\text{isolated, 3 dimensional})}} = \frac{\eta_{(\text{swarm, 2 dimensional})}}{\eta_{(\text{isolated, 2 dimensional})}} \quad (\text{VII-12})
\]

where \(\eta\) represents one of the properties of the flow patterns which we have already considered.

VII-3-1 Estimate of Properties

If the horizontal array of bubbles just studied may be taken to represent a swarm of bubbles, then the throughflow and recirculation coefficients and the normalized height of the recirculation region for a three dimensional swarm may be estimated. The values of these properties were obtained by use of Tables 1-3 and eqn. (12) and are shown in Figs. 7-9.

Use of eqn (12) is a definite improvement over simply assuming that the properties of a bubble within a swarm are the same as those of an isolated bubble. Most of the trends found in Figs. 7-9 compare with the isolated three dimensional bubble as expected.
Ratio of Bubble to Emulsion Velocities ($\alpha$)

FIGURE VII-7 - Throughflow Coefficient in a Three Dimensional Swarm of Slow Bubbles
Recirculation Coefficient (β)

Ratio of Bubble to Emulsion Velocities (α)

Fig.(VII-8) Recirculation Coefficient in a Three Dimensional Swarm of Slow Bubbles
Figure VII-9 - Normalized Recirculation Height in a Three Dimensional Swarm of Slow Bubbles
In Fig. 7, the existence of neighboring bubbles tends increasingly to reduce the amount of throughflow as a/b increases. The variation of the throughflow with the bubble to emulsion velocities ratio has the same tendencies as with the isolated bubble: as $\alpha$ increases, throughflow decreases. The effect of the neighboring bubbles increases as the cloud increases, as is seen from how the curves fall off most rapidly as $\alpha > \alpha_c$. The effect of "three dimensionalizing" is to increase the throughflow of the two dimensional case.

In Fig. 8 the recirculation coefficient estimate for the three dimensional swarm also shows results as expected. Eqn. (12) and the expressions in Table 2 were used. For $\alpha \rightarrow 1$, the recirculation region becomes infinitely large, but still much smaller than that corresponding to a single bubble as the region in which it may expand is restricted. On the other limit, as $\alpha \rightarrow 0$, the reverse occurs. The recirculation region in the single bubble case becomes negligible. This is not the case for groups of bubbles where the streamfunction indicates that recirculation regions may exist as $\alpha = 0$. The latter is only a result of the approximation involved in the evaluation of the streamfunction of the array of cylindrical bubbles. There the pressure field was assumed to be created by a row of sources, one at the center of each bubble. Due to interactions between the sources, the surfaces of constant pressure are not circular, and hence the boundary of the bubble is represented by a deformed circle elongated in the horizontal direction. The extra region covered by this elongated circle is calculated erroneously as the recirculation region.
Fortunately, except for high ratios of $a/b$, the additional region is negligible compared to the bubble volume. Also, $\alpha = 0$ is a hypothetical case which does not occur as bubbles are never stationary in a fluidized bed. Again, wall effects are accentuated when $\alpha \gtrsim \alpha_c$.

Similar comments may be made of Fig. 9. That $\chi$ should be evaluated as finite for $\alpha \rightarrow 0$ is again the result of the inadequacy of the representation of the constant pressure surface at the bubble boundaries by sources. Again the value of $\chi$ at this limit is negligible for $a/b$ ratios which are not too close to 1. An unexpected result is observed as $\alpha \rightarrow 1$. The only curve which tends to infinity as $\alpha \rightarrow 1$ is that of the isolated bubble; all other cases tend to zero. This is physically unacceptable as the volume of the recirculation region must grow in order to occupy all the medium. Mathematically, the combination suggested by eqn. (12) for the normalized height of the three dimensional swarm tends to zero proportionally to $-(1 - \alpha)^{1/6} \ln(1 - \alpha)$ as $\alpha \rightarrow 1$. For a fixed $a/b$ ratio, the only direction in which the recirculation region may grow is the vertical. So, as $\beta$ increases as $-\ln(1 - \alpha)$ as $\alpha \rightarrow 1$, the normalized height should also increase as $-\ln(1 - \alpha)$. Physically, however, this problem should not occur in a swarm as bubbles above the one considered would restrict both the recirculation volume and height from growing to infinity. The normalized recirculation height generally becomes larger than the bubble radius once $\alpha > 0.8$, approximately.
VII-3-2 Modifications

In order to simplify the computation of the above properties of three dimensional bubble swarms, the dotted lines in Figs. 5 and 6 have been taken as approximations of the ratios

\[ C = \frac{\beta (\text{isolated, 2 dimensions})}{\beta (\text{isolated, 3 dimensions})} = 2.2633 \alpha + 1.3333 \quad \text{(VII-13a)} \]

\[ C = \frac{\chi (\text{isolated, 2 dimensions})}{\chi (\text{isolated, 3 dimensions})} = 2 \quad \text{(VII-13b)} \]

Calculations of the corresponding properties of the three dimensional swarms depends only on the appropriate coefficients for the horizontal array of cylindrical bubbles in Tables (2) and (3) divided by the coefficients in eqns. (13). The results are shown in Figs. 10 and 11 and the corresponding analytical expressions are in Table (4). Notice their similarity with Figs. 8 and 9, respectively and that the normalized height of the recirculation does tend to grow indefinitely as \( \alpha \) nears one. This, as was argued above, is what should be expected physically.

As in Figs. 8 and 9, the error at the lower limit of \( \alpha \) has persisted. However, the values of \( \beta \) and \( \chi \) are negligible except for \( a/b \) ratios approaching one. Also, in practice bubbles are not stationary in a fluidized, so \( \alpha \neq 0 \) always.

The error involved in using these approximations for \( C_\beta \) and \( C_\chi \) are less than 2% for the recirculation coefficient and less than 20% for the normalized height for \( \alpha \leq 0.95 \). The trend of the normalized
Fig. (VII-10) Modified Recirculation Coefficient in a Three Dimensional Swarm of Bubbles.
Fig. (VII-11) Modified Normalized Recirculation Height for a Three Dimensional Swarm of Bubbles
Table VII-4

Properties of three dimensional swarms of bubbles

<table>
<thead>
<tr>
<th>Property</th>
<th>Condition</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Throughflow coefficient</td>
<td>$\alpha &lt; \alpha_c$</td>
<td>[ \frac{3}{2} \frac{(1+\alpha/2)^{2/3}}{(1+\alpha)^{1/2}(1-\alpha)^{1/6}} [ \nu \arccos \left( 1- \frac{T}{y} \right) + (2\nu T - T^2)^{1/2} ] ]</td>
</tr>
<tr>
<td></td>
<td>$\alpha \geq \alpha_c$</td>
<td>[ \frac{3}{2} \frac{(1+\alpha)^{2/3}(1-\alpha)^{5/6}}{(1+\alpha)^{1/2}} \frac{b}{a} ]</td>
</tr>
<tr>
<td>Recirculation coefficient</td>
<td></td>
<td>[ \frac{1}{2C_\beta} \left[ \frac{1}{\pi a} \int_{y_c}^{y_Q} \frac{y}{\sqrt{d+y^2-1}} , dy + \frac{y_c}{a} \sqrt{a^2-y_c^2} - 2 \arctan \left( \frac{\sqrt{a^2-y_c^2}}{y_c} \right) \right] ]</td>
</tr>
<tr>
<td>Normalized recirculation height ($\chi$)</td>
<td>$\gamma &lt; 3$</td>
<td>[ \frac{1}{C_\chi} \max \left[ \frac{\chi}{a}(y_1), \frac{\chi}{a}(y_c) \right] ]</td>
</tr>
<tr>
<td></td>
<td>$\gamma \geq 3$</td>
<td>[ \frac{1}{\pi C_\chi \frac{a}{b}} \ln(\gamma - \sqrt{2\gamma - 2}) ]</td>
</tr>
</tbody>
</table>

Note: $\beta, C, \chi, a, b$ are defined by eqns(13), $d$ is eqn(18), $x/a$ is eqn(7c) when $x^2 = a^2 - y^2$, $y_0$ is defined by eqn(11b), $y_1$ is root of eqn(11a), $y_c = a - \sqrt{2}$ is defined by eqn(5).
height coefficient $C$ for $0.95 < \alpha < 1$ does not apply as has already been discussed. Consequently this approximation seems satisfactory for the whole range of $\alpha$.

VII-3-3 Discussion of Bubble Swarms

It should be noted that the effects studied here are only due to bubbles at the horizontal level, and so the influence of bubbles above and below have not been considered. Before the recirculation regions of two vertically positioned bubbles overlap, it is probable that the influence on the flow pattern of horizontally neighboring bubbles may be considerably larger. However, it may be speculated that when the recirculation regions of two vertical bubbles overlap gas may flow from one bubble to another and so the vertical effect would then become important. Passage of gas from one fast bubble to another has been noticed by some authors, such as Werther [4].

Another problem is how one should relate the parameter $a/b$ of the horizontal string of two dimensional bubbles with a three dimensional swarm of bubbles. It is suggested that $a/b$ be taken as the ratio between bubble diameter $(2a)$ and average distances between bubbles in the horizontal and vertical directions are $2b$, then the bubble fraction may be expressed as

$$\delta = \frac{\pi}{6} \left( \frac{a}{b} \right)^{1/3}$$

An alternative would be to assume a height $l$ between bubbles proportional to the bubble radius. For example, Darton et al [5] assume that $l = 2^{4/3} a$. Then

$$\delta = \frac{2^{2/3}}{3} \left( \frac{a}{b} \right)^2$$
\[ \delta = \frac{2^{2/3}}{3} \left( \frac{a}{b} \right)^2 \]

if the unit cell corresponding to a bubble was assumed to be a circular cylinder of height \( l \) and radius \( b \).

As \( \alpha \) increases, the total gas flow in a bed will be hindered by the reduction of the throughflow coefficient and the increase in recirculation volume which reduces the region where emulsion gas travelling at a higher velocity may occupy. The increasing velocity of the bubble enhances the general flow, but may not be sufficient to overcome the loss of flow mentioned. This effect of \( \alpha \) on total flow in the bed will be considered in the next section.

The properties of three dimensional swarms are summarized in Table (4). A computer program that calculates these properties is given in Appendix VII-1.

**VII-4 THE EFFECTIVE THROUGHFLOW COEFFICIENT**

In section (II-3-1) the mass conservation equation was given for the fast bubble case neglecting the cloud fraction. In this section the mass conservation equation for slow bubbles, including both the recirculation and throughflow coefficients will be considered. This may be written as

\[
\frac{[\text{Gas through emulsion}]}{[\text{volume of gas contained in rising bubble region}]} = u_o = u_f \left[ 1 - (1 + \beta) \delta \right] + \frac{\delta(1 + \beta \epsilon_{mf})u_b + m \delta u_{mf}}{[\text{flow through bubble}]} (VII-14)
\]
The particles in the emulsion have been assumed stationary, so the wake and the difference between the interstitial velocity at minimum fluidization and the emulsion velocity were neglected. It is more convenient to combine the recirculation with the throughflow in a factor $M$, which would yield a simplified mass conservation equation:

$$u_o = u_f (1-\delta) + \delta u_b + M \delta u_{mf} \quad (VII-15)$$

Applying eqn. (II-5) and requiring equations (14) and (15) to be equivalent:

$$M = m + (\alpha + \kappa - 1) \beta \quad (VII-16)$$

where

$$\kappa = \left( \frac{u_o}{u_{mf}} - 1 \right) \epsilon_{mf} \quad (VII-17)$$

Figs. 12-15 are graphs representing $M$, the effective throughflow coefficient, at different values of $\kappa$ and use of the equations in Table (4).

The case $\kappa = 0$ in Fig. 12, which implies incipient fluidization, is unexpected. The effective throughflow drops sharply and in a few occasions becomes negative as the volume occupied by the recirculation region deprives the flow within the emulsion. For $\alpha + 1, M + 0$ as then both emulsion and bubble gas are travelling at the same velocities and the eqn. (14) is correct even with $M = 0$. 
Fig. (VII-12) Equivalent Throughflow Coefficient for a Three Dimensional Swarm of Bubbles ($\kappa=0$)
Fig. (VII-13) Equivalent Throughflow Coefficient for a Three Dimensional Swarm of Bubbles (κ = 0.5)
Fig. (VII-14) Equivalent Throughflow Coefficient for a Three Dimensional Swarm of Bubbles ($\kappa = 1.0$)
Fig. (VII-15) Equivalent Throughflow Coefficient for a Three Dimensional Swarm of Bubbles (κ = 3.0)
For higher values of $\kappa$, given in Figs. 13-15, a common pattern is observed. $M$ increases with $\kappa$, i.e. with excess superficial velocity. For low values of $a/b (<0.5)$, $M$ lies approximately between 2 and 3 for the lower range of $\alpha (<0.8)$. According to Glicksman et al. [6], experimental data yields effective throughput coefficients which have a value of about 3. However, the data used probably has rather high $a/b > 0.5$. On the higher range of $\alpha$, $M$ is shown to increase indefinitely. This has no physical meaning as the recirculation region cannot extend indefinitely although the model used does allow this to occur. If the height and volume of the recirculation region are restricted to grow to a certain maximum, $M$ would reach a finite value at $\alpha = 1$.

VII-5 CONCLUSION

Three of the flow properties for slow bubbles have been investigated, resulting in an estimate of them for a swarm of three dimensional bubbles. The throughput coefficient, recirculation coefficient, and normalized height are given in Table (4) as a function of the parameters $\alpha$ and $a/b$, using the simplifications suggested by eqns (13). An effective throughput coefficient was defined. It was found that for small $a/b$ ratios and in most of the range of $\alpha$ this coefficient was close to 3, which is approximately the value found experimentally according to Glicksman et al [6].
VII-6 NOMENCLATURE FOR CHAPTER VII AND ITS APPENDICES

a - bubble radius [L]
b - half-distance between neighboring bubbles in a horizontal array [L]
\(C_\beta, C_x\) - ratios defined in eqns. (13)
d - defined by eqn. (8)
h - height of the recirculation region [L]
\(\ell\) - vertical distance between bubbles [L]
m - throughflow coefficient
M - effective throughflow coefficient
r - radial coordinate [L]
\(r_o\) - radial coordinate of the stagnation point between the recirculation region and the emulsion [L]
s - \(\left[(1-\alpha)/(1+\alpha)\right]^{1/2}\)
T - Defined by eqn. (2)
u_b, u_{br} - absolute bubble velocity, and bubble velocity relative to particles at infinity [L/T]
u_f, u_{mf} - interstitial and superficial velocities at minimum fluidization [L/T]
w - potential for horizontal array of cylindrical bubbles [L^2/T]
x, y - cartesian coordinates with origin at the bubble origin [L]
x_c, y_c - coordinates of point of intersection between the limiting
stream surface and the bubble interface [L]

\( (x_o, y_o) \) - coordinates of the stagnation point on the limiting stream surface [L]

\( z = re^{i\theta} \) [L]

Greek Symbols

\( \alpha \) - \( u_{br}/u_f \), the bubble to emulsion velocity ratio

\( \alpha_c \) - critical bubble to emulsion velocity ratio, defined by eqn. (4)

\( \beta \) - recirculation coefficient

\( \gamma \) - \( = \nu T \)

\( \delta \) - bubble to bed volume fraction

\( \epsilon_{mf} \) - voidage at minimum fluidization

\( \eta \) - property of the flow pattern

\( \theta \) - angular coordinate

\( \kappa \) - defined in eqn. (17)

\( \nu \) - defined in eqn. (5)

\( \phi \) - \([ (1-\alpha)/(1+\alpha/2) ]^{2/3} (y/a)^2\)

\( \psi, \psi_o \) - streamfunction and value of the limiting streamsurface [L^2/T] or [L^3/T]

\( \chi \) - normalized recirculation height
References of Chapter VII and its Appendices


CHAPTER VIII

CONCLUSIONS AND RECOMMENDATIONS

VIII-1 Introduction Throughout the work presented in this thesis one problem recurrently surfaces: there is not sufficient knowledge of the fluid mechanics of a fluidized bed to predict quantitatively the mass transfer processes with reasonable confidence. To understand the mass transfer phenomena in greater detail the flow processes related to bubbles must be known. This work has hinged on present models of bubbles which could prove inadequate for present design requirements. Consequently, the main effort in future work should be in understanding the fluid mechanics.

Other conclusions and recommendations will be viewed separately, depending on the topic studied.

VIII-2 Mass transfer from fast bubbles to emulsion The models presented in the literature are still at a primitive level. The experimental work in bubble to emulsion gas mass transfer done up to now is inadequate as few of the parameters required in the calculation of a mass transfer coefficient are measured experimentally; so many assumptions must be made. Recently fewer but more careful work has been made. This should continue.

A plot of all available experimental bubble to emulsion mass transfer data in minimally fluidized beds was presented in fig. (III-15). Certain trends are found; namely: a) that the Davidson and Harrison
mass transfer model approximates experimental results for the larger particles (>500µ), where the particle Reynolds number is high, and when the bubble diameter approaches the bed diameter; (b) the Chiba and Kobayashi and Kunii and Levenspiel models are conservative estimates for small particle systems (<500µ); and (c) Davidson and Harrison's model is also adequate for very small particle systems (<80µ).

Effects rarely considered up to now should be relevant in enhancing the transfer process. Some of these are: (a) the interactions and perturbations of neighboring bubbles (wake shedding and cloud overlapping being those of greatest importance), (b) the dispersion of gas across the cloud being higher than molecular diffusion due to the presence of particles which enhance gas mixing at moderate particle Reynolds numbers (>5) and (c) laminar wakes attached to the particles at Reₚ larger than about 5.

Careful experimental data is needed. Experiments may be directed to finding radial dispersion coefficients in the emulsion phase, further work with three dimensional bubble mass transfer with measurement of all parameters involved including both emulsion and bubble concentrations, investigations of the fluid mechanics of bubble interaction and eventually the mass exchange in a bed with swarms of bubbles.

From the data available it is apparent that the dependence of the mass transfer coefficient \( K_{be} \) is less dependent on bubble diameter than theoretically predicted \( (K_{be} \sim 1/d_b^{0.25-1}) \). This may be related to variation of the stability of the bubble with size.
For present use it is recommended that the equations given in Table (IV-1) be used for mass transfer aspects of design.

VIII-3 Radial gas dispersion by slow bubbles  The model presented in Chapter V for overall radial dispersion in a slow bubbling bed should be conservative. Comparison with experiments made with fast bubbling beds indicate that probably radial mixing will be higher in slow bubbling beds than in fast bubbling beds.

Theoretically, small bubbles have shown to be good mixing points. There have been no experiments measuring radial dispersion in slow bubbling beds. This is what is foremostly needed. Also, as before, a better knowledge of the fluid mechanical processes is required. Confirmation that bubbles are good mixing points is also needed. In the modeling, the bubble's motion, the recirculation of gas around the bubbles and accounting for bubble to bubble interactions is required.

VIII-4 Particle to fluid mass and heat transfer in fixed and fluidized beds  The experimental results for packed beds which indicate that Sherwood and Nusselt numbers for individual particles approach zero at low Reynolds numbers are caused by improperly accounting for or omitting important phenomena at low Reynolds number. These include flow channeling due to high voidage along the walls of the test vessel, and inaccuracies in the concentration or temperature measurements. Analyses such as Galloway and Nelson's penetration theory are inappropriate at small Reynolds numbers and, moreover, when used with a reasonable definition of $Sh$ do not indicate a fall of $Sh$ to zero.
At low Reynolds numbers, the actual Nusselt and Sherwood number for individual particles in a fixed or fluidized bed approach a value equal or greater than unity.

When the overall heat or mass transfer of a fixed or fluidized bed is to be determined at low Reynolds numbers, a careful consideration of channeling and axial diffusion must be included. Channeling may be prevented in a fixed bed by having ratios of particle to vessel diameters of about or over 1000. A good knowledge of the bubble to emulsion mass transfer coefficients is also a requirement for experiments in fluidized beds.

VIII-5 Throughflow and recirculation properties of isolated and swarms of slow bubbles  The throughflow coefficient, the recirculation coefficient and normalized height were calculated for two and three dimensional isolated bubbles and for a two dimensional swarm of bubbles by use of the Davidson and Harrison bubble model assumptions. It is possible to obtain with this information reasonably good estimates of these properties for bubbles in a three dimensional swarm. A combination of them yields the effective throughflow coefficient $M$ for a bubble in a three dimensional swarm, for use in the mass conservation equation for the fluid mechanical model for a fluidized bed. A combination of eqn. (VIII-16) and Table (VI-4) define $M$ in terms of the ratio of the bubble diameter to distance between neighboring bubbles $(a/b)$, the ratio of bubble to interstitial velocities $(\alpha = u_b / u_f)$ and the ratio of the excess superficial velocity to the interstitial velocity $\kappa = (u_o - u_{mf}) / u_f$. The effective throughflow coefficient, for values of $a/b$ which do not approach 1, has
a relatively constant value (close to 3) except for values of $\alpha$ approaching 1 where $M$ increases indefinitely.

The approximations involved in the modeling of the two dimensional swarm are not too good for values of $a/b$ approaching 1. Consequently, the effective throughput coefficient for bubbles in a three dimensional swarm is not too accurate in this upper range of $a/b$. Future modeling efforts should consider this problem. Also, the effect on these properties of neighboring bubbles in relative positions to one another different from horizontal should be studied further.
APPENDIX II-I

Requirement for Plug Flow in the Emulsion. The dispersive model which will be assumed for the plug flow in the emulsion has been used by Baerns et al (1963) and Hiraki et al (1968) for the entire fluidized bed:

\[
u \frac{\partial C}{\partial z} = \frac{D_r}{r} \left( \frac{\partial}{\partial r} r \frac{\partial C}{\partial r} \right) + D_a \frac{\partial^2 C}{\partial z^2} + S(r,z)
\]

where \(D_a\) and \(D_r\) are axial and radial diffusion coefficients. This is the dispersed plug flow model as described by Bischoff and Levenspiel (1962). Note that for flat radial concentrations within the emulsion the source term may not be radially dependent. This source term describes the exchange between the bubble and emulsion phases. So if the bubbles are uniformly distributed at each height, then \(s(r,z) = s(r)\). In that case this term will not affect the radial distribution and from now on will be neglected. The resultant equation has been solved for a continuous point source by Klinkenburg et al (1953) and is

\[
\frac{C}{C_a} = 1 + \sum \frac{1}{q_i} \exp \left[ \left(1 - \sqrt{1 + x_i^2} \right) \frac{z}{2R} \frac{D_r}{D_a} \right] \frac{J_r(j_i, i \frac{r}{R})}{J_0(j_i, i)}
\]

where

\[
q_i = \sqrt{\frac{1}{4} + x_i^2} \quad \text{and} \quad x_i^2 = \frac{D_a}{u^2 R^2} j_i, i^2
\]

\(j_i, i\) being the \(i\)'th root of \(J_1(x) = 0\), and \(C_a\) the concentration of the axis of the bed.

Impermeability at the walls \((r=R)\) was also assumed. If the
argument of the exponential is large, then it may be neglected and \( C = C_a(z) \). This may occur only if

\[
\left( \frac{\frac{D}{r}}{\frac{u}{R} \frac{D}{r}} \right)^2 \left( \frac{z}{R} \frac{D}{r} \right)^{1/2} \gg 1
\]

or

\[
\frac{D}{u_o R} \frac{2 \frac{D}{r}}{L^2} \gg \ll 1
\]

where \( L \) is a characteristic height of the bed and the roots of \( J_0(z) \) have been disregarded as they are all larger than one.
Concentration Profiles Within the Cloud and Surrounding Emulsion

The method described by Boussinesq (1906) will be used here. The coordinates of the bubble system of Figure (1) \((r, \theta)\) are transformed into the coordinates \((\phi', \psi)\), the stream function \(\psi\) modified potential function \(\phi'\). The relation between the coordinates is

\[
\psi = - u_f (\alpha - 1) \left( 1 - \frac{r c^3}{r^3} \right) \frac{r^2 \sin^2 \theta}{2}
\]

\[
\phi' = \frac{2}{3} u_f (\alpha - 1) \left( 1 + \frac{r c^3}{2r^3} \right) r^3 \left( 1 - \frac{3}{2} \cos \theta + \frac{\cos^3 \theta}{2} \right)
\]

Following Boussinesq, it is assumed that diffusion is only important in the transverse direction to the flow. Then

\[
\frac{\partial C}{\partial \phi'} = D \frac{\partial^2 C}{\partial \psi^2}
\]

The boundary condition is chosen at the cloud-emulsion interface, where the concentration \(C_{co}\) is assumed to be constant. It is shown in Appendix (IV-3) that if the bubble and emulsion concentrations are assumed constant, then \(C_{co}\) is their average. Hence, if the rate of mass transfer is not rapid, the above assumption is adequate. This is in contradiction to Drinkenburg's (1970) assumption that the bubble's concentration is everywhere constant, and as a result found that the concentration \(C_{co}\) did not vary with angle \(\theta\). The assumption is not altogether adequate as the gas reentering the bubble after being depleted of the species of interest may not be at the same concentration as the gas emerging from the bubble. For large clouds both
Different Bubble Coordinates

Figure AII-2-1
boundary conditions are compatible except near \( \theta = \pi \). The boundary condition is then taken as

\[ C = C_{co} \text{ at } \psi = 0 \]

The solution is

\[ \frac{C - C_{co}}{C_b - C_{co}} = \text{erf} \left( - \frac{\psi}{\sqrt{4D\phi'}} \right) \quad \psi < 0 \]

or in terms of the coordinates \((r, \theta)\)

\[ \frac{C - C_{co}}{C_b - C_{co}} = \text{erf} \left\{ \left[ 3 (\alpha - 1) \frac{r_c}{D} \frac{u_f}{u_c} \right]^{1/2} \frac{(1 - 1/x^3)x^2 \sin^2 \theta}{4[(2x^3 + 1)(1 - 3/2 \cos \theta + \cos^3 \theta)^{1/2}]} \right\} \]

where \( x = r/r_c \)

Note that \( C_b \) is really the concentration of the gas at the point \((r_b, 0)\). The solution for two cases is given in Figures (2) and (3). It should be remarked that the profiles in the emulsion are symmetrical about the point \( C = C_{co} \), \( r = r_c \) of the graphs in the immediate neighborhood of the bubble. The complete solution for \( \phi' > 0, \psi > 0 \) is

\[ \frac{C - C_{co}}{C_e - C_{co}} = \text{erf} \left( \frac{\psi}{\sqrt{4D\phi'}} \right) - H(\phi' - a) \]

\[ \cdot \frac{\psi}{\sqrt{4D\phi'}} \int_{0}^{\frac{a}{\phi' - a}} e^{-\eta^2} \text{erf} c \left[ \eta \left( \frac{a}{\phi' - a} \right) \right]^{1/2} d\eta \]
Modified Concentrations

\[ \frac{C-C_{co}}{C_b-C_{co}} \]

\[ \theta = \frac{5}{6} \pi \]

\[ \frac{u_f r_b}{D_e} = 10 \]

\[ \alpha = 2 \]

modified radius \( \frac{r - r_b}{r_c - r_b} \)

Concentration profiles within the cloud at different angles

Figure AII-2-2
Concentration profiles within the cloud at different angles.

Figure AII-2-3
where \( H(x) \) is the Heaviside function and \( a = \phi'(r_c, \pi) \). The second term of the right hand side is incorporated because the boundary layer at \( \psi = 0 \) for \( \phi' > 0 \) (beyond the cloud-emulsion interface) is

\[
\frac{3}{\sigma} \left( \frac{C - C_{co}}{c_e - c_{co}} \right) \left. \bigg|_{\theta = \pi, r > r_c} \right. - \frac{3}{\sigma \psi} \left( \frac{C - C_{co}}{c_e - c_{co}} \right) \left. \bigg|_{\psi = 0, \phi' > a} \right. = 0
\]

due to the symmetry in the emulsion phase about the vertical axis of the bubble.
Solutions to the Differential Equations Relating Emulsion Concentration Profiles to the Exchange Coefficients.

In all cases it is necessary to use the tracer conservation equation:

\[ \frac{F_{c}C_{e} + F_{b}C_{b} + F_{c}C_{c}}{o} = F_{o}C_{o} \quad \text{AII-3-1} \]

where the subscripts \( b, c, e \) refer to the bubble, cloud and emulsion phases. The subscript \( o \) refers to the entering bubble properties.

Model 1) When the bubble volume is assumed constant and the mass transfer resistances are defined at the bubble-cloud and cloud-emulsion interfaces Eqn. (II-13) has to be solved. \( C_{e} \) is substituted in Eqn. (II-13b) from the tracer conservation Eqn. (II-1).

The resulting expression is differentiated and linearly combined with Eqn. (II-13b), \( n_{o} \) is dependent on \( C_{e} \), in such a way that \( C_{b} \) and \( dC_{b} \) are eliminated by use of Eqn. (II-13a). The resulting equation is

\[ \frac{d^{2}C_{c}}{dh^{2}} + \frac{a}{dh} \frac{dC_{c}}{dh} + bC_{c} = d \]

where

\[
\begin{align*}
a &= \left[ \frac{1}{(C_{c} + F_{w})} + 1 \right] \frac{K_{be}}{u_{b}} + \frac{K_{ce}(1 + F_{e}C_{e})}{u_{b}(C_{c} + F_{w})} \\
b &= \frac{K_{bc}K_{ce}}{(C_{c} + F_{w})C_{e}u_{b}} \left( \frac{F_{T}}{F_{e}} \right)^{2}
\end{align*}
\]
\[
d = \frac{K_{bc} K_{cc}}{(f_c + f_c)} \frac{F_C e}{F_e} \frac{C_o}{C_0}
\]

where \( f_c \) is the cloud fraction.

The general solutions to these equations are

\[
C_b = A_1 e^{\lambda_1 h} + A_2 e^{\lambda_2 h} + \frac{F_C e}{F_T} C_0
\]

\[
C_c = B_1 e^{\lambda_1 h} + B_2 e^{\lambda_2 h} + \frac{F_C e}{F_T} C_0
\]

where \( \lambda_1, \lambda_2 = \frac{-a \pm \sqrt{a^2 - 4b}}{2} \)

In order to satisfy Eqn. (II-13a) the following relation between the coefficients must hold.

\[
\lambda_1 A_i = \frac{K_{bc}}{u_b} (B_i - A_i) \quad i = 1, 2
\]

or

\[
B_i = A_i \lambda_i + \frac{K_{bc}}{u_b} \frac{K_{bc}}{u_b} \quad i = 1, 2
\]

In order to satisfy boundary conditions

\[
A_1 + A_2 + \frac{F_C e}{F_T} C_0 = C_{bo}
\]

\[
\left( \frac{\lambda_1 + \frac{K_{bc}}{u_b}}{K_{bc}/u_b} \right) + \frac{\lambda_2 + \frac{K_{bc}}{u_b}}{K_{bc}/u_b} + \frac{F_C e}{F_T} = C_{co}
\]

\[
C_b = C_b(0), \quad C_{co} = C_c(0)
\]
Solving for $A_1$ and $A_2$

$$A_i = \left[ \frac{F_0 C_o}{F_T} (\lambda_i + \frac{K_{bc}}{u_b} - 1) - C_{bo} (\lambda_i + \frac{K_{bc}}{u_b}) + C_o \right] \frac{K_{bc}/u_b}{\lambda_j - \lambda_i} \text{ AII-3-2}$$

where $i = 1, 2$ and $j = 3 - i$

By use of Eqn. (1)

$$C_e = C_1 e^{\lambda h} + C_2 e^{2h} + \frac{F_0 C_o}{F_T}$$

with

$$C_1 = -\frac{F_e A_i + F A_i}{F_e}$$

Model 2) When bubble volume is assumed variable, but only one interface resistance is assumed, the tracer conservation equation becomes

$$F_b C_b + F_e C_e = F_T C_\infty$$

where $F_T$ is the total volumetric gas flow and $C_\infty$ the average volumetric concentration at a bed cross-section of interest and substituting $C_e$ into Eqn. (II-14)

$$\frac{d V_b C_b}{dT} + a V_b C_b = b$$

where

$$a(h) = \frac{F_T K_{be}}{F_e u_b} \quad \text{and} \quad b(h) = \frac{K_{be} F_T C_\infty}{F_e u_b}$$

By multiplying both sides by

$$e(h) = \exp \int_0^h a(x) \, dx$$
it is possible to integrate this equation directly. The solution is

\[ C_b = \left[ \int_{h_0}^{h} b(x)e(x) \, dx + V_{bo}C_{bo} \right] \over e(h) - V_b \]

where the subscript \( o \) denotes properties taken at the height of the injector. The emulsion concentration is given by

\[ C_e = \frac{F_T}{F_e} \frac{C_\infty}{C_e} - \frac{F_T}{F_e} \frac{F_b}{V_b} e(h) \int_{h_0}^{h} b(x)e(x) \, dx + \]

\[ + \frac{V_{bo}}{V_b e(h)} \left[ -\frac{F_T}{F_e} + \frac{F_T}{F_b} \frac{F_{bo}}{F_e} C_{eo} \right] \]

When \( C_\infty = 0 \), as for Latham and Potter's (1970) work, then it is reduced to,

\[ C_e = \frac{F_T}{F_e} \frac{F_{bo}}{F_b} \frac{F_{eo}}{F_e} \frac{V_{bo}}{V_b} C_{eo} \exp \left\{ \int_{h_0}^{h} \frac{K_e}{u_b} \frac{F_T}{F_e} \, dx \right\} \]

Model 3) When constant volume and only one interfacial resistance is assumed, the solution to Eqn. (II-15) is obtained by differentiating the tracer conservation equation and substituting for \( C_b \)

\[ \frac{d}{dx} \frac{C_e}{C_e} = -\frac{F_T}{F_b} \frac{F_{bo}}{u_b} \left[ \frac{F_T}{F_e} \frac{C_e}{C_e} - \frac{F_T}{F_b} \right] \]

\[ \frac{d}{dn} \ln \left( C_e - \frac{F_T}{F_e} C_{\infty} \right) = -\frac{K_e}{u_b} \frac{F_T}{F_e} \]
So the slope of this modified logarithmic concentration profile determines the exchange coefficient, if $F_n$, $F_e$ and $u_b$ are known. The last two are sometimes hard to determine.
APPENDIX III-1

Estimate of Error Incurred by Assuming Only Emulsion Gas is Sampled.

If a probe samples a fraction \( a \) of gas from the bubble phase during a measurement in the emulsion phase, then the actual concentration measured will be

\[
\frac{C_{\text{ave}}}{C_\infty} = a \frac{C_b}{C_\infty} + (1 - a) \frac{C_e}{C_\infty}
\]

AIII-1-1

where \( C_\infty = \frac{F_o C_\infty}{F_T} \) is the concentration at \( x = \infty \).

If the models of (II-3) assumed is that with one resistance to mass exchange, the Eqn. (II-15) is integrated to give

\[
\frac{C_e - C_\infty}{C_{e0} - C_\infty} = \frac{F_{e0}}{F_{b0}} \frac{F_b}{F_e} \exp \left( - \int \frac{h}{h_o} \frac{K_{be} F_T}{u_b F_e} \, dx \right)
\]

and by use of the tracer conservation Eqn (II-1)

\[
\frac{C_b - C_\infty}{C_{b0} - C_\infty} = \exp \left( - \int \frac{h}{h_o} \frac{K_{be} F_T}{u_b F_e} \, dx \right)
\]

Allowance for variation in properties is made because, as seen in Appendix II-3 the variable bubble model may be reduced to the form of the constant volume bubble by a change in the interchange coefficient, \( K_{be} \).

By incorporating these two equations into Eqn. (1) and applying the tracer conservation Eqn. (II-1), a relationship between the concentra-
tion of the two phases and that actually measured is found to be

\[
\frac{C_{\text{ave}} - C_{\infty}}{C_{e_0} - C_{\infty}} = \frac{F_{e_0}}{F_{b_0}} \frac{F_{e_0}}{F_{b_0}} - a(1 + \frac{F_b}{F_e}) \exp - \int_{h_0}^{h} \frac{K_{\text{be}} F_T}{u_b F_e} \, dx
\]

For the case of constant bubble properties the logarithm of the above expression should be considered. Then

\[
\ln \left| \frac{C_{\text{ave}} - C_{\infty}}{C_{e_0} - C_{\infty}} \right| = \ln \left| 1 - \frac{a F_T}{F_e} \right| - \frac{K_{\text{be}} F_T}{u_b F_e} (h - h_0)
\]

So on a plot of the logarithm of the actually measured modified concentration profile, the error will be \( \ln \left| a \frac{F_T}{F_e} - 1 \right| \). Note that if \( a \) were the same at each sampling, the slope of the profile remains unchanged, as Latham and Potter (1970) have already pointed out. Fryer and Potter (1972, 1975) have assumed that \( a = \delta \), but this really should depend not on the time fraction the sampling probe is exposed to the bubbles, but on the volume of gas sampled from this phase, which may be different.

In order to make an approximate estimate of the error due to the fluctuation in the fraction \( a \), it will be assumed that \( a \ll \delta \). If \( \frac{F_T}{F_e} \delta \ll 1 \), the error \( \epsilon \) may be estimated as

\[
|\epsilon| \leq \delta \frac{F_T}{F_e}
\]

which gives an error bar around \( C_b \). The error in the slope will then
be approximately

$$|\varepsilon_s| = \frac{2 \delta F_T/F_e}{h_{\text{max}} - h_{\text{min}}}$$

where the properties in the numerator are averages and $h_{\text{min}}$ and $h_{\text{max}}$ are the minimum and maximum heights at which the gas was sampled. So the error in the exchange coefficient due to this is approximated by

$$|\varepsilon_k| < \frac{2 \delta/u_b}{h_{\text{max}} - h_{\text{min}}}$$

where $u_b$ is now an average too. Notice that the error is small for most of the experiments encountered.
Tables of Available Bubble Mass Transfer Coefficient Data

The data for the bubble mass transfer coefficient which has appeared in the literature is tabulated for isolated two and three dimensional bubbles. The data was read from tables or from graphs in papers and modified into a standard form. Other data of possible influence on the exchange is also included. When it was not available, a zero was entered. No attempt was made to differentiate data taken in the slugging regime from those in the bubbling, if the author has not made this clear. Data from papers working exclusively in the slugging regime has not been included. This is the data used in the attempted correlations.
<table>
<thead>
<tr>
<th>Investigator</th>
<th>$u_{mf}$ (cm/s)</th>
<th>$d_p$ (µ)</th>
<th>$e_{mf}$</th>
<th>D (cm$^2$/s)</th>
<th>$d_b$ (cm)</th>
<th>$\frac{1}{u_{mf}d_b}$</th>
<th>$\frac{D_{eg}^{2/4}}{K_{be}d_b}$</th>
<th>$\frac{1}{u_{mf}}$</th>
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<td>DAVIES AND RICHARDSON (1966)</td>
<td>0.1426 00</td>
<td>0.5172 00</td>
<td>0.1463 00</td>
<td>0.529E 01</td>
<td>0.383E 01</td>
<td>0.312E 02</td>
<td>0.379E 02</td>
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<td>and $u_{mf}$ derived from</td>
<td></td>
<td></td>
<td></td>
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$u_{mf}$ is superficial velocity at minimum bubbling - at which bed was operated

### PEREIRA AND CALDERBAUGH (1975)

- **0.575** E 00
- **96**
- **0.557** E 00
- **0.704** E 00

### WALKER (1975)

- **0.165** E 01
- **97**
- **0.550** E 00
- **0.205** E 00

---

*Data taken from Chiba and Kobayashi (1970)*
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<th>Investigator</th>
<th>( u_{mf} ) (cm/s)</th>
<th>( d_0 ) (( \mu ))</th>
<th>( e_{mf} )</th>
<th>( D ) (cm(^2)/s)</th>
<th>( d_b ) (cm)</th>
<th>( \frac{1}{u_{mf}} ) ( \frac{D^2}{d_b} )</th>
<th>( K_{be} ) ( \frac{d_b}{u_{mf}} )</th>
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Note: The data is derived from equations (2.20) and (2.22) of Sherwood et al. (1976).
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-23-
The two dimensional mass exchange bubble.

AIII-3.1 The Davidson and Harrison model. The derivation follows that given by Davidson and Harrison (1963) for the three dimensional case. It is first noticed that throughflow is given by

\[ Q_c = 2[\psi(r_c, \frac{\pi}{2}) - \psi(r_b, \frac{\pi}{2})] \varepsilon_{mf} \]

where the stream function

\[ \psi(r, \theta) = u_f(\alpha - 1) \left(1 - \frac{r_c^2}{r^2}\right) r \sin \theta \]

Substitution yields

\[ Q_c = 2 \ u_{mf} \ d_b \]

The mass exchange due to convection is

\[ N_c = 2 \ u_{mf} \ d_b \ (C_b - C_e) \]

The diffusive part is then obtained by a component mass balance on the bubble-cloud interface, on the bubble side. The gas flow is assumed to be parallel to this interface and diffusion to take place only in the direction perpendicular to the flow. Taking \( y \) and \( \theta \) as the coordinates as shown in Fig. (1), the component mass balance near the wall becomes

\[ D \ \frac{\partial^2 C}{\partial y^2} \ dy \ d\theta = \ \frac{u_y}{r_b} \ \frac{\partial C}{\partial \theta} \ d\theta \ dy \quad \text{AIII-3-1} \]

Approximating the stream function near the wall as \( \psi = u_y y \) where \( u_y \) is assumed independent of \( y \) near the wall. Then, equation (1) may be written as
\[
\frac{D}{u_\theta} \frac{\partial^2 C}{\partial \psi^2} = \frac{u_\theta}{r_B} \frac{\partial C}{\partial \theta} \quad \text{AIII-3-2}
\]

Defining the variable \( \phi \) by \( \frac{\partial \phi}{\partial \theta} = r_B u_\theta \), equation (2) is rewritten as

\[
D \frac{\partial^2 C}{\partial \psi^2} = \frac{\partial C}{\partial \phi} \quad \text{near } \psi = 0. \quad \text{AIII-3-3}
\]

The concentrations at the interface \((\psi = 0)\) is \(C_b\), and at infinity \(C_e\), i.e. at \((\psi = \infty)\), are assumed to be constant. The solution becomes

\[
\frac{C - C_e}{C_b - C_e} = \text{erfc} \left( \frac{\psi}{2(D\phi)^{1/2}} \right) \quad \text{AIII-3-4}
\]

The exchange for a bubble subtending the angle 2\(\theta\), is

\[
N_c = -2 D \int_0^\theta \frac{1}{2} \frac{\partial C}{\partial \psi} \bigg|_{\psi = 0} r_B \, d\theta
\]

Change of variable yields

\[
N_c = -2 D \int_0^{\phi(\theta_1)} \frac{3C}{\partial \psi} \bigg|_{\psi = 0} \, d\phi
\]

which is integrated to give

\[
N_c = \frac{\dot{\theta}}{4} \left( \frac{D\phi(\theta_1)}{\pi} \right)^{1/2} \left( C_b - C_e \right) \quad \text{AIII-3-5}
\]

Note that \( \phi(\theta) = \int_0^\theta r_B u_\theta \, d\theta \) from equation (3). Also \( u_\theta \) is found by assuming Bernoulli's integral to be applicable. In that case

\[
u_\theta = \left[ 2 g r_B (1 - \cos \theta) \right]^{1/2} \quad \text{AIII-3-6}
\]
Model of a Two Dimensional Bubble

Figure AIII-3-1
Substituting this into the expression for $\phi$ and integrating

$$\phi (\theta) = 2 \left( r_B g \right)^{1/2} \left( 1 - \cos \theta \right)$$

Substituting into equation (5)

$$N_d = \frac{2}{\pi^{1/2}} \left( D d_B^3 g \right)^{1/2} \left( 1 - \cos \theta_1 \right)^{1/2} \left( C_b - C_e \right)$$

As the bubble subtends on angle $2\theta_1$ of a circle, it would be convenient to write the mass exchange as a function of the equivalent bubble diameter $d_b$ (i.e., diameter of a spherical bubble with the same volume).

A short calculation yields

$$d_b = \left( \frac{\theta_1 - \cos \theta_1 \sin \theta_1}{\pi} \right)^{1/2} d_B$$

This expression is substituted into equation (6) and added to the convective expression for interchange (where $d_B$ is replaced for $d_b$)

$$N_T = \left[ 2 \frac{u_{mf}}{d_p} + 3.78 \frac{D d_b^3 g}{(2 \theta_1 - \sin 2\theta)^{3/8}} \right] \left( D d_B^3 g \right)^{1/2} \left( 1 - \cos \theta_1 \right)^{1/2} \left( C_b - C_e \right)$$

The exchange coefficient per unit bubble volume is

$$K_{be} = 2.55 \frac{u_{mf} + 4.81}{d_b} \left( \frac{D g}{d_b^{5/2}} \right)^{1/2}$$

Chavarie and Grace (1972, 1976) and Walker (1975) have assumed that $\theta_1 = \pi$, which yields
\[ K_{be} = 2.55 \frac{u_{mf}}{d_b} + 2.42 \left( \frac{D \frac{g}{5/2}}{d_b} \right)^{1/2} \]

If Murray's (1965a) suggestion that \( \theta_1 = 7\pi/9 \) is applied

\[ K_{be} = 2.55 \frac{u_{mf}}{d_b} + 2.48 \left( \frac{D \frac{g}{5/2}}{d_b} \right)^{1/2} \]

If Davidson and Harrison's (1963) suggestion for the three dimensional bubble is used, i.e. \( \theta_1 = 50^\circ \), then

\[ K_{be} = 2.55 \frac{u_{mf}}{d_b} + 5.33 \left( \frac{D \frac{g}{5/2}}{d_b} \right)^{1/2} \]

A-III-3.2 The Chiba and Kobayashi Model

From Collins (1969), the complex potential for Murray's (1965b), two dimensional bubble is

\[ w(z) = u_F [(1-\alpha)z - r_b^{-2} \alpha/z - r_b^{-3/4}z^2] \]

The stream function is therefore given by

\[ \psi = \text{Im}(w) = u_F \left[ (1-\alpha) + \frac{a r_b^{-2}}{r^2} \frac{\alpha^{-1}}{4} \sin \theta + \frac{a^3}{4 \pi} \sin 2\theta \right] \]

As in the Davidson and Harrison case, the diffusion equation is

\[ \frac{u_b}{r_c} \frac{\partial C}{\partial \theta} = D \frac{\partial^2 C}{\partial r^2} \]

Note that in this case the diffusion is considered at the cloud-
emulsion interface. Also

\[ \frac{\partial \phi}{\partial \theta} \bigg|_{r_c} = r_c u_\theta \bigg|_{r_c} \]

\[ u_\theta \bigg|_{r_c} = -\frac{\partial \psi}{\partial r} \bigg|_{r_c} = -u_f [(1-\alpha) - \alpha r_b^3 - \frac{r_b^3}{r_c^2} - \frac{r_b^3}{r_c^3} \cos \theta] \sin \theta \]

\( \psi \) is linearized around \( r = r_c \), \( \psi = -u_\theta \bigg|_{r_c} \frac{r_c(r - r_c)}{r_c} \)

Under these conditions equation (8) may be written as

\[ D \frac{\partial^2 C}{\partial \psi^2} = \frac{\partial C}{\partial \psi} \]

Assume that at the concentrations at the cloud emulsion interface (\( \psi = 0 \))
and very far away in the emulsion (\( \psi = \infty \)) are constant and equal to \( C_c \) and
\( C_e \) respectively. The concentration profile is given by equation 7.16. The total exchange is given by

\[ N_c = -\int_0^{\theta} D \varepsilon \frac{\partial C}{\partial r} \bigg|_{r=r_c} r_c d\theta \]

As before, the integral may be written as a function of \( \psi \) and \( \phi \). After integrating

\[ N_c = 4 \left( \frac{D\phi(\theta_1)}{\pi} \right)^{1/2} \varepsilon \frac{\partial \psi}{\partial r} (C_b - C_e) \]
where
\[
\phi(\theta) = \int_{r_c}^{r_b} u_\theta \, d\theta = -u_f [(1-\alpha)r_c - \frac{r_b}{r_c} \alpha] (1-\cos\theta) + \frac{u_f r_b^3}{2 r_c^2} \sin^2 \theta
\]

The exchange coefficient per unit bubble area is given by
\[
K_a = \frac{4}{\pi^{3/2}} \varepsilon_{mf} \left(\frac{D}{d_c}\right)^{1/2} \left[ (1-\alpha) \frac{d_c}{2} - \frac{d_b}{d_c} \alpha \right] (1-\cos \theta) + \\
+ \frac{d_b^3}{d_c^3} \frac{\sin^2 \theta}{4}^{1/2}
\]

It will be further assumed that \( \theta = \pi \). Then, by substitution of the relation of cloud to bubble diameter \( (d_b/d_c)^2 = \frac{\alpha}{\alpha-1} \) and \( \alpha = u_{br}/u_f \), the exchange coefficient becomes
\[
K_a = \frac{25}{4\pi^3} \varepsilon_{mf} \left(\frac{D}{d_c}\right)^{1/2} \left(\frac{\alpha-1}{\alpha}\right)^{1/2}
\]

The exchange coefficient per unit (bubble + cloud) volume is
\[
K_{ce} = \frac{4K_c}{d_c} = 4.06 \varepsilon_{mf} \left(\frac{D}{d_c}\right)^{1/2} \left(\frac{\alpha-1}{\alpha}\right)^{1/2}
\]

The exchange per unit bubble volume is
\[
K_{be} = \frac{4.06}{1-F_w} \varepsilon_{mf} \left(\frac{D}{d_b}\right)^{1/2} \left(\frac{\alpha-1}{\alpha}\right)^{1/4}
\]
(III) The Kunii and Levenspiel model. This has been reported by Chavarie and Grace (1972, 1976) as

\[
K_{ce} = 4.51 \left( \frac{D_e \epsilon_{mf} u_b}{d_b^3} \right)^{1/2} \epsilon_{mf} \quad D < D_e < D
\]

Comparison of two dimensional models with data. The three models are compared with data from a number of different experiments in Fig. (2). The curves which have been plotted are:

Davidson and Harrison

\[
\pi_2 = 2.55 + 2.48 \pi_1
\]

Kunii and Levenspiel

\[
\pi_2' = 2.55 + 2.48 \pi_1 \\
\pi_2'' = 2.17 \epsilon_{mf}^{1/2} \pi
\]

and

\[
1/\pi = 1/\pi_2' + 1/\pi_2''
\]

Chiba and Kobayashi

\[
\pi_1 = 1.95 \epsilon_{mf} \Pi_1
\]

where

\[
\pi_1 = \frac{1}{u_{mf}} \left( \frac{D_\epsilon \epsilon_{mf}^{1/2} d_b^{1/2}}{d_b^{1/2}} \right)^{1/2} \quad \Pi_2 = \frac{K_{be} d_b}{u_{mf}}
\]

and

\[
u_b = 0.48 (g d_b)^{1/2}, \text{ the latter being suggested by Grace}
\]
and Chavarie (1972). In order to draw the curves in the last two models it was assumed that \( \varepsilon_{mf} = 0.5 \).

The conclusions to be drawn from this graph are the same as for the three dimensional bubble case of Fig. (III-15). The bubble diffusion models do not seem to be entirely applicable.
Plot of Two Dimensional Bubble to Emulsion Exchange Data for Minimally Fluidized Beds

Figure AIII-3-2
Determination of the Average Particle Reynolds Number in the Cloud Region

Assuming that the Davidson and Harrison bubble model is correct, the average Re \(_p\) in the cloud may be found as follows.

The speed of the fluid within the emulsion with respect to the moving particles may be found from the stream function given by Rowe (1971):

\[ \psi_f = -u_f \left[ 1 + 2 \frac{r_b^3}{r} \right] \frac{r^2 \sin^2 \theta}{2} \]

where \((r, \theta, \phi)\) is a spherical coordinate system with origin at the center of the bubble of radius \(r_b\). Calculation of the radial and angular velocities yields

\[ u_r = u_f \left[ 1 + 2 \frac{r_b^3}{r} \right] \cos \theta \quad \text{AIV-1-1} \]
\[ u_\theta = -u_f \left[ 1 - \frac{r^3}{r_b^3} \right] \sin \theta \quad \text{AIV-1-2} \]

The average velocity is given by

\[ u_{\text{ave}} = \frac{\int_{r_b}^{r_c} \int_{\theta_o}^{\pi} \sqrt{u_r^2 + u_f^2} \ 2\pi r \ \sin \theta \ d\theta \ dr}{\int_{r_b}^{r_c} \int_{\theta_o}^{\pi} 2\pi r \ \sin \theta \ d\theta \ dr} \]

where \(r_c\) is the cloud radius. Rearranging and making change of variables,
\[ \rho = \frac{r}{r_b} \]

\[ u_{ave} = \frac{3}{2} \frac{u_f}{(\rho_c^3 - 1)} \left( \int_1^{\rho_c} \sqrt{(1 + \frac{2}{\rho^3})^2 \cos^2\theta + (1 - \frac{1}{\rho^3}) \sin^2\theta} \right) \times \rho^2 \sin\theta \, d\theta \, dp \]

\[ = \frac{3}{2} \frac{u_f}{(\rho_c^3 - 1)} \int_1^{\rho_c} \left[ (\rho^3 - 1)^2 \arg\sin h \frac{\sqrt{6\rho^3 + 3}}{\rho^{3\rho^3 + 3}} \rho^{3\rho^3 + 3} + \rho^{3+2} \rho \right] dp \]

where \( \rho_c = \frac{r_c}{r_b} \).

An approximation to the above integral may be given in two parts.

a) For \( r_b < r_c < 2r_b \), the integrand may be expanded in a Taylor series around \( \rho = 1.5 \). This was then integrated and then divided by \( (\rho_c^3 - 1) \) and again expanded around \( \rho = 1.5 \). The solution is given by

\[ \frac{u_{ave}}{u_f} \approx -0.3898 \frac{r_c^3}{r_b^3} + 2.1303 \frac{r_c}{r_b} - 4.0557 \frac{r_c}{r_b} + 3.7590 \]

\[ 1 < \frac{r_c}{r_b} < 2 \]

The expansions used always had an error which was less than 2%. The region of highest error is around \( \frac{r_c}{r_b} = 1 \), where the error is 3.75%, theoretically \( \frac{u_{ave}}{u_f} = \frac{3}{2} \) at \( \frac{r_c}{r_b} = 1 \) while the approximation gives 1.444.
b) For \( \frac{r_c}{r_b} \geq 2 \), integration from 2 to \( \frac{r_c}{r_b} \) is required as the rest of the integration has just been dealt with. For \( \rho > 2 \), it is noted that, within one percent, the integrand is approximated by \( 2 \rho^2 \) so

\[
\frac{u_{ave}}{u_f} = \frac{\rho_c^3}{\rho_f^3} - 8 \frac{\rho_c^3}{\rho_f^3} - 1 + \frac{7.3528}{\rho_c^3 - 1}
\]

\[
\frac{u_{ave}}{u_f} = \left( \frac{r_c}{r_b} \right)^3 - \frac{0.6472}{l} \frac{r_c}{r_b} \geq 2
\]

This expression is virtually equal to 1. As a result, the average particle Reynolds number within the cloud may be approximated by

\[
Re_p = \frac{u_{ave} d_p}{\nu} =
\]

\[
\left\{ \begin{array}{l}
-0.3898 \frac{r_c}{r_b}^3 + 2.1303 \frac{r_c}{r_b}^2 - 4.0577 \frac{r_c}{r_b} + 3.7590 \times Re_F \\
= \left( \frac{r_c}{r_b} \right)^3 - 0.6472 \times Re_F \quad \text{if } 1 < \frac{r_c}{r_b} < z.
\end{array} \right.
\]

\[
= \left( \frac{r_c}{r_b} \right)^3 - 1 \times Re_F \quad \text{if } \frac{r_c}{r_b} \geq 2.
\]

Where \( Re_F \) is the particle Reynolds number based on the interstitial gas velocity at minimum fluidization \( u_f \). A plot is given in Fig. (1).

It should be stressed that this value may be wrong, as the validity of all the bubble models are questionable near the bubbles themselves as Jackson (1971) has indicated.
Dimensional Analysis of the Exchange Coefficient

There are a number of equations which must be considered. As the gas exchange takes place at the interstitial level, the behavior of the fluid at this level must be taken into account. The equations describing the average behavior of the fluid-particle system, in the form Jackson (1971) has published, reportedly summarized the implicit and explicit assumptions of the most important models published up to date, namely Davidson and Harrison's (1963) Murray's (1965) and Jackson's (1964). They describe the average flow around a number of particles. Consequently it is necessary to return to the more fundamental Navier Stokes equations. It is proposed here to use the mentioned fluid-particle system equations which have as characteristic properties the large scale properties of the bed; and the local fluid behavior will be described by the Navier Stokes equations, with characteristic properties being imposed by the local flow and a characteristic interstitial length.

The equations describing the system's behavior are

(a) continuity equations. The flows are assumed to be incompressible and consequently no information is obtained from them.

(b) average momentum equation. For the average behavior of the particles

\[ \rho_s (1-\varepsilon) \nabla (\nabla \cdot \mathbf{v}) + \rho_s g (1-\varepsilon) - \beta(\varepsilon) (\mathbf{U} - \mathbf{v}) = 0 \]

and of the fluid
\[ \nabla p + f(\varepsilon) (\vec{U} - \vec{V}) = 0 \]

where

\[ \rho_s = \text{particle density, assumed constant} \]
\[ \varepsilon = \text{local average void fraction} \]
\[ \vec{V} = \text{local average particle velocity vector} \]
\[ \beta = \text{stress function on particles} \]
\[ \vec{U} = \text{local average fluid velocity vector} \]
\[ \nabla p = \text{local average pressure gradient} \]

The above equations are from Jackson (1971).

Taking the frame of reference in which the bubble is stationary, the two phase flow model would suggest the \( \vec{U} = (u_f - u_b) \vec{i} \) and \( \vec{V} = -u_b \vec{i} \) far from the bubble. The characteristic length is \( d_b \), the bubble diameter, \( \varepsilon_{mf} \) is a characteristic value of \( \varepsilon \) and \( \nabla p - \rho_s g (1 - \varepsilon_{mf}) \) as reported by Kunii and Levenspiel (1968). So

\[ (1 - \varepsilon) (\vec{V}' \vec{V}') + \frac{g d_b}{u_b} (1 - \varepsilon)' - \frac{f(\varepsilon_{mf}) d_b}{\rho_s u_b^2 (1 - \varepsilon_{mf})} [ (u_f - u_b) \vec{U}' + u_b \vec{V} ] = 0 \]

and

\[ \nabla p' + \frac{\varepsilon_{mf}}{\rho_s g (1 - \varepsilon_{mf})} \varepsilon' [ (u_f - u_b) \vec{U}' + u_b \vec{V} ] = 0 \]

where

\[ (1 - \varepsilon)' = \frac{(1 - \varepsilon)}{1 - \varepsilon_{mf}} \]
\[ p' = \frac{p - p_a}{\rho_s g (1 - \varepsilon_{mf})} \]
\[ \vec{V}' = -\frac{\vec{V}}{U_b}, \quad \vec{U}' = \frac{U}{u_f - u_b} \]

\[ \vec{V}' = d_b \vec{V}, \quad \vec{P}' = \frac{\vec{P}(\varepsilon)}{\vec{P}(\varepsilon_{mf})} \]

so \( U', V', \) and \( P' \) are all functions of

\[ \frac{g}{u_b^2}, \quad \frac{\beta(\varepsilon_{mf})}{\rho_s} \frac{d_b}{u_b^2} \frac{u_f - u_b}{1 - \varepsilon_{mf}} \quad \frac{\varepsilon(\varepsilon_{mf})}{\rho_s U_b (1 - \varepsilon_{mf})} \quad \text{and} \quad \frac{R}{d_b} \]

the dimensionless vector indicating the position of the region considered.

c) the Navier Stokes equation. For an incompressible fluid with constant viscosity,

\[ \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \vec{u} = -\nabla P + \nu \nabla^2 \vec{u} \]

where \( \vec{U} \) is the fluid's real velocity and \( \rho_f \) is the fluid density.

We will now consider the fluid in the frame where the particle is on the average stationary. The characteristic gas velocity is \( \vec{U} - \vec{V} \); the characteristic length is \( d_p \), the particle diameter, the characteristic time is that due to bubble rising in the neighborhood, \( d_b/u_b \); and the pressure, the pressure \( \vec{p} - p_a \). Then

\[ \frac{U_b}{d_b} \frac{d_p}{\vec{U} - \vec{V}} \frac{\partial \vec{u}'}{\partial t} + \left( \vec{u}' \cdot \nabla' \right) \vec{u}' = \frac{g d_p}{(\vec{U} - \vec{V})^2} - \frac{(\vec{p} - p_a)}{\rho_f (\vec{U} - \vec{V})^2} \nabla' p' \]

\[ + \frac{\vec{v}''^2 u'}{d_p (\vec{U} - \vec{V})} \]
where
\[ u' = \frac{u}{U-V}, \quad \tau' = \frac{u}{d_b} \tau \]
\[ \psi'' = \frac{d_y}{d_p}, \quad \rho' = \frac{P - P_a}{P - P_a} \]

so \( U' \) and \( p' \) are functions of
\[ \frac{u_b}{U-V} \frac{d_p}{d_b} \left( \frac{g}{d_p} \right)^2, \quad \frac{P - P_a}{\rho(U-V)^2}, \quad \frac{\psi}{d_p(U-V)} \text{ and } \frac{\rho}{d_p} \]

where \( \rho/d_p \) is the dimensionless position vector.

Note that instead of using \( U-V \), it will be sufficient to characterize the velocity by \( u_x \), as this is the value achieved far away.

Also \( P-P_a \) will be replaced by \( P_s (1-\varepsilon_{mf}) g d_b \), for the same reasons. Then, in the bubble frame
\[ u' = \frac{u}{u_f} = f \left( \frac{u_b}{u_f} \frac{d_p}{d_b} \right), \quad \frac{g}{d_p} \left( \frac{1-\varepsilon_{mf}}{\rho_f u_f^2} \right), \quad \frac{\psi}{d_p u_f} \frac{g}{d_b}, \quad \frac{\rho}{d_p} \]
\[ \frac{\varepsilon_{mf}}{\rho_s u_b(1-\varepsilon_{mf})}, \quad \frac{\varepsilon_{mf}}{\rho_s u_b(1-\varepsilon_{mf})}, \quad \frac{R}{d_b}, \quad \frac{R}{d_p} \]

It should be noted that the characteristics length really is that of the interstices between the particles. This depends on \( d_p, \varepsilon_{mf}, \) and \( \theta \), the shape factor. Nevertheless, \( d_p \), is probably a good measure. Also the interstitial velocity will be a function of the motion of the particles.
and should enter the above equation as a boundary condition. Finally, the dimensionless parameters may be reduced to:

\[ u' = \frac{d_p}{d_b}, \frac{u_f}{u_b}, \frac{g}{u_f^2}, \frac{\rho_s g (1 - \varepsilon_{mf}) d_b}{\rho_f u_f^2}, \frac{u_f}{v}, \frac{R}{d_p}, \frac{R}{d_b}, \text{boundary conditions} \]

d) The diffusion equation. At the interstitial level

\[ \frac{C}{t} + (\mathbf{u} \cdot \nabla) C = D \nabla^2 C \]

the characteristic properties are those given in (c), besides the concentration difference, \( c_1 - c_2 = \Delta c \), between the two phases being considered. Then,

\[ \frac{u_b}{u_f} \frac{d_p}{d_b} \frac{\partial c'}{\partial t} + (\mathbf{u}' \cdot \nabla') C' = \frac{D}{u_f} \frac{d}{d_p} \nabla''^2 C' \]

where \( c' = c/\Delta c \). Clear \( c' \) is a function of

\[ \frac{D}{u_f} \frac{d}{d_p}, \frac{u_b}{u_f} \frac{d_p}{d_f}, u' \text{ and } \frac{r}{d_p} \]

Now, the net mass transfer out of nondeformable control surface around the cloud travelling at velocity \( u_b \).
\[
\frac{dN}{dt} = \int \left[ D \nabla \cdot C + (u - v) C \right] \cdot \mathbf{n} \, dA_c
\]

where \( N \) is the number of moles of tracer contained within the control surface, \( \mathbf{n} \) the normal to it and \( \nabla \cdot C \) the gradient with respect to a local frame of reference traveling at \( u_b \). Noting that the control surface is taken either at the bubble-cloud or cloud-emulsion interface and that \( \frac{d_c}{d_b} = h(u_b/u_f) \)

\[
\frac{1}{d_b^2 u_b \Delta C'} \frac{dN}{dt} = g \frac{D}{u_f d_p} \frac{u_b}{u_f} \frac{d_p}{d_b} \frac{\rho_s g (1 - \varepsilon_{mf}) d_b}{\rho_f u_f^2} \frac{g d_b}{u_b^2}
\]

\[
\frac{u_f d_p}{v} \frac{\beta(\varepsilon_{mf}) d_b}{\rho_s d_b (1 - \varepsilon_{mf})}, \text{ boundary conditions).}
\]

The relation is

\[
\frac{dN}{dt} = -K_{12} C
\]

Equating these two expressions

\[
\frac{K_{12}}{u_b d_b^2} = \frac{g \frac{D}{u_f d_p} \frac{u_b}{u_f} \frac{d_p}{d_b} \frac{g d_b}{u_b^2} \frac{\rho_s g (1 - \varepsilon_{mf}) d_b}{\rho_f u_f^2}}{u_f d_p} \frac{\beta(\varepsilon_{mf}) d_b}{\rho_s d_b (1 - \varepsilon_{mf})}, \text{ boundary conditions).}
\]

AIV-2-1
Fast Bubble Gas Exchange Model Proposed

There are a number of bubble-emulsion gas exchange models which have appeared in the literature. They are meant to be used at low particle Reynolds numbers, where Stokes' flow applies and possibly molecular diffusion is an important mechanism for mass transfer. However, in a fluidized bed combustor the particles' Reynolds numbers at the exchange site (i.e. the cloud) is typically higher, in a region where Stokes' flow does not apply but viscous forces are still important. The following is an attempt to extend the models into the region of interest and indicate other mechanisms which may also apply.

It will be assumed that Davies's law still applies and that Davidson and Harrison's bubble model is adequate. The bubble and cloud is assumed to be perfectly mixed and the mechanisms are dependent on interactions between the cloud and emulsion. The particles play an important role.

**Interfacial Mass Transfer**

(i) The effective diffusion coefficient. Exchange may occur at the limit between the cloud and emulsion gas flows. At these higher Reynolds numbers, molecular diffusion should lose importance. This occurs in packed beds, where at particle Reynolds numbers larger than about 1, the effective diffusion equation starts to depend on the gas velocity relative to the particle as Wilhelm (1962) indicates. In fluidized beds dispersion in the emulsion phase, when the particles are not in relative motion between each other, is probably well described by the fixed bed model. However, when
the particles are in motion relative to each other, the basic structure of a packed bed exists no longer, making the analogy less valid. It is probable that the relative particle motion should enhance the dispersion, but in the writer's knowledge no theory or experiments have been proposed on this topic. Edwards and Richardson (1968) have speculated that some very high axial diffusion coefficients they found was due to motion of particles within their packed bed, but the evidence is not conclusive. Due to the present lack of knowledge, which was recognized earlier by Kunii and Levenspiel (1968), it is proposed to use de Ligny's (1970) equation,

\[
\frac{1}{Pe_{a,r}} = \frac{\gamma}{Pe_p} + \frac{\lambda_{a,r}}{1 + \frac{C}{Pe_p}} \tag{1}
\]

where

\[
Pe_{a,r} = \frac{u_d}{Da_r}, \quad Pe_p = \frac{u_d}{D}
\]

\(\lambda_{a,r}\) and \(C\) being constants and \(\gamma = 1/T\) where \(T\) is the tortuosity \((T = \sqrt{2})\)

Table (1) gives the values of the constants for gas according to de Ligny. The subscripts \(a\) and \(r\) refer to axial and radial conditions. It should be noted that there remains a lot of scatter in the data, and the \(Pe_{a,r}\) are usually plotted against \(Re_p\) instead of \(Pe_p\) as the dependence on the molecular diffusion coefficient of \(Pe_{a,r}\) has not been entirely accepted. Sherwood et al. (1976) affirm the probability that no unique correlation between \(Pe\), \(Re\) and \(Sc\) exist to explain the dispersion phenomena.
<table>
<thead>
<tr>
<th>Particles</th>
<th>Spherical</th>
<th>Irregular</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \lambda_d ) (longitudinal dispersion)</td>
<td>0.7</td>
<td>4</td>
</tr>
<tr>
<td>( \lambda_r ) (radial dispersion)</td>
<td>0.12</td>
<td>0.12</td>
</tr>
<tr>
<td>( C ) (longitudinal dispersion)</td>
<td>5.8</td>
<td>5.1</td>
</tr>
<tr>
<td>( C ) (radial dispersion)</td>
<td>78 ( \pm ) 20</td>
<td></td>
</tr>
</tbody>
</table>

An average dispersion coefficient must be taken for flow at the interface. Setting \( r = r_c \) in equations (II-1,2) the average angular and radial velocities with respect to the particles are found by averaging over the cloud-emulsion interface.

\[
\overline{U}_r = \frac{3\alpha U_f}{2(\alpha+2)}
\]

\[
\overline{U}_\beta = \frac{3\pi U_f}{4(\alpha+2)}
\]

where

\[
\left( \frac{r_c}{r_b} \right)^3 = \frac{\alpha+2}{\alpha-1}, \quad \alpha = \frac{U_{br}}{U_f}
\]

The magnitude \( U_{ax} \) and the angle \( \beta \) with respect to the normal surface of the cloud of these velocities are given by (see Figure (1))

\[
\overline{U}_{ax} = \frac{3 U_f}{2\pi(\alpha+2)} \sqrt{1 + (\pi/2\alpha)^2}
\]

\( \beta = \arctan \frac{\pi}{2\alpha} \)
The average dispersion coefficient normal to the wall will be given

\[ D_e = D_r \sin \beta + D_a \cos \beta \]  

(2)

where \( D_r \) and \( D_a \) are functions of \( D, \overline{U_{ax}} \) and \( d_p \) given by equation (1) \( (\overline{U_{ax}} = U_e) \).

(I) The mass exchange coefficient. In the following, the effective diffusion coefficient \( D_e \) will be assumed constant.

(a) For large clouds \( [D_e C / U_f R_b] << 1 \) when the cloud is large, it is not possible to assume that its concentration \( C_c \) is constant. However, the distribution of the concentration within the bubble has little effect on the final distribution at the cloud-emulsion interface. The streamflow by the Davidson and Harrison stream function \( \psi \) and potential function \( \phi \).

Following Bousinesq (1906) the axisymmetric flow is transformed into the \( (\psi, \phi') \) plane, where

\[ \phi' = \int r^2 \sin^2 \theta \, d\phi \]

being the potential function.

The cloud-emulsion interface \( (r = r_b) \) has been transformed to \( \psi = 0, \, 0 < \phi' < 2(\alpha + 2)U_f R_b^2 \). The emulsion phase lies in the positive semiplane and the cloud and bubble phases in the negative \( \psi \) semiplane. The bubble cloud interface is represented by
Velocity at cloud emulsion interface

Figure AVR-3-1
\[ \psi = -\frac{3}{2} \frac{u_f}{r_b} \sin^2 \theta \]

\[ \phi' = u_f \alpha r_b^3 \left[ 1 - \frac{\cos \theta}{2} (3 - \cos^2 \theta) \right] \]

\[ 0 < \theta < \pi \]

For the presence of the bubble void to be negligible, the thickness of the boundary layer thickness \( \delta_c \) where gas is depleted at the cloud emulsion emulsion interface must be much smaller than the cloud thickness, i.e.,

\[ \left( \frac{D \phi'}{\psi} \right)^{1/2} \ll 1 \text{ at } r = r_b \]

Thence

\[ \frac{D \alpha}{u_f r_b} \ll \frac{4}{9} \sim 1 \]

Defining the concentration at \( r = r_c \) as \( C_c(\phi') \), of the distant emulsion as \( C_e \) and within the bubble as \( C_{b\infty} \), from Boussinesq's paper

\[ \gamma_1 = \frac{C - C_e}{C_c(\phi') - C_e} = \frac{\sqrt{2/\pi}}{\int_0^\infty f(\phi' - \frac{\psi^2}{2D\phi'\eta^2}) e^{-\eta^2/2} \, d\eta} \quad (4a) \]

and

\[ \gamma_2 = \frac{C - C_b}{C_c(\phi') - C_b} = \frac{\sqrt{2/\pi}}{\int_0^\infty f(\phi' - \frac{\psi^2}{2D\phi'\eta^2}) e^{-\eta^2/2} \, d\eta} \quad (4b) \]

where

\[ f(\phi') = \begin{cases} 1 & 0 \leq \phi' \leq 2u_f(\alpha+2) r_b^3 \\ 0 & \text{elsewhere} \end{cases} \]
A species balance at $\psi = 0$ yields

$$
(C_c (\phi') - C_e) \frac{\partial \gamma_1}{\partial \psi} \bigg|_{\psi=0} = (C_b (\phi') - C_b) \frac{\partial \gamma_2}{\partial \psi} \bigg|_{\psi=0}
$$

But from Equation (3)

$$
\frac{\partial \gamma_1}{\partial \psi} \bigg|_{\psi=0} = \frac{\partial \gamma_2}{\partial \psi} \bigg|_{\psi=0}
$$

Substituting into equation (4) and adding terms, it is determined that

$$
C_c (\phi') = C_{co} = \frac{C_b + C_e}{2}
$$

Thence, the total rate of bubble to emulsion exchange is

$$
N_{bc} = \frac{C_b - C_e}{2} 2 \sqrt{\frac{D_e}{\pi \varepsilon_{mf}}} \int_0^1 \sqrt{2U_f (\alpha + 2)r_b^3} d\eta
$$

Converting to the standard exchange coefficient per unit bubble volume

$$
K_{be} = 3.39 \left( \frac{D_e (U_{br} + 2U_f)}{d_b^3} \right)^{1/2}
$$

When the large cloud assumption is used, the cloud volume should not be neglected in the bubble to emulsion mass exchange calculations of the bed.
For Small Clouds \( \frac{D e}{U_f r_b} \ll 1 \)

The bubble model of Davidson and Harrison is derived from a potential flow, the total mass exchange coefficient is given by Boussinesq (1906) which may be written as

\[
K_G = 4 \sqrt{\pi D e} \varepsilon_{mf}^2 \frac{U_f (\alpha - 1)}{U_f (\alpha - 1)(\phi'_1(\pi))^{1/2}}
\]  

(7)

where

\[
\phi'_1(\theta) = \frac{\int_0^\theta \frac{r_c^2 \sin \theta}{U_f (\alpha - 1)} d \phi}{U_f (\alpha - 1)}
\]

and

\[
\phi(r_c, \theta) = -\frac{3}{2} \frac{U_f (\alpha - 1)}{r_c} \cos \theta
\]

so

\[
\phi_1(\theta) = -\frac{3}{2} \frac{r_c^2}{U_f (\alpha - 1)} \int_0^\theta (1 - \cos^2 \theta) d \cos \theta
\]

Performing the integration and setting \( \theta = \pi \)

\[
\phi_1(\pi) = 2 \frac{r_c^2}{U_f (\alpha - 1)}
\]

Substituting into equation (7) and observing that \( K = K_{be} \frac{\pi d_b}{6} \)

\[
K_{be,d} = 6.77 \left( \frac{D e \varepsilon_{mf}^2 U_f (\alpha + 2)}{d_b^3} \right)^{1/2}
\]
Particle with a Conical Wake

Figure AIV-3-2
If a wake fraction \( w \) is defined as its volume divided by the particle's volume,

\[
w = \left( V_{OBD} + V_{BCD} - V_{OBED} \right) \times \left( \frac{\pi d}{6} \right) -1
\]

Observing that \( \cos \theta = \frac{1}{2s/d_p + 1} \) and combining the above equations the following simple relation for \( w \) is obtained

\[
w = \frac{(s/d_p)^2}{2s/d_p + 1} \tag{8}
\]

Taneda (1956) plotted \( s/d_p \) versus \( Re_p \) for the case of a single sphere in an infinite medium. The resulting curve may be expressed by

\[
s/d_p = 0.629 \ln \left( \frac{Re_p}{24} \right) \quad 24 < Re_p < 130
\]

Substituting into equation (8)

\[
w = \frac{0.40 \ln (Re_p/24)^2}{1.26 \ln (Re_p/24 + 1)} \quad (Re_p > 24) \tag{8a}
\]

ii) Particle through the cloud. From Davidson and Harrison's model, it is assumed that the particle stream function is given by

\[
\psi_p(r, \theta) = U_br \left( 1 - \frac{r_b}{r^3} \right) \frac{r^2 \sin^2 \theta}{2} \tag{9}
\]
The total particle volume flow rate $Q_p$ through the cloud is given by

$$Q_p = 2\pi [\psi_r(r_c, \pi/2) - \psi_r(r_b, \pi/2)](1 - \varepsilon_{mf})$$

Substituting equation (9) into the above,

$$Q_p = Q_g \frac{1 - \varepsilon_{mf}}{\varepsilon_{mf}} \frac{\alpha}{[(\alpha+2)(\alpha-1)]^{1/3}}$$

(10)

where

$$Q_g = 3\pi r_b^3 U_{mf}$$

is the total bubble flow rate.

The total gas flow rate $Q_w$ in the form of a wake is given by

$$Q_w = Q_p w$$

(11)

Substituting equations (8a-l) the exchange is obtained

$$Q_w = Q_g \frac{\alpha}{[(\alpha+2)(\alpha-1)]^{1/3}} \frac{1 - \varepsilon_{mf}}{\varepsilon_{mf}} \frac{0.40[\ln(Re_p/24)]^2}{1.26 \ln (Re_p/24) + 1}$$

The exchange coefficient is therefore (24 < $Re_p$ < 130)

$$K_{be,w} = \frac{Q}{2d_b} \frac{U_{mf}}{[(\alpha+2)(\alpha-1)]^{1/3}} \frac{1 - \varepsilon_{mf}}{\varepsilon_{mf}} \frac{0.40[\ln(Re_p/24)]^2}{1.26 \ln (Re_p/24) + 1}$$

An estimate of the particle Reynolds number is obtained by use of $U_{ax}$ as the typical velocity. It should be observed that as the wake should not exceed the interstitial volume it is additionally required that
\[ w < \frac{\varepsilon_{mf}}{1 - \varepsilon_{mf}} \]
Exchange due to growth. None of the models considered account for exchange due to bubble growth. Consequently, when using Eqn.

\[
\frac{u_b}{b} \frac{dV_b}{dh} b = K_{bc} V_b (C_b - C_c)
\]

The increase of moles of the tracer in the bubble due to this increase of size is neglected. The relation will be derived in the frame of reference of the bubble. A bubble has interchange by three different ways.

(1) by simple exchange of gas with the emulsion as described by most of the models.

(2) by addition of gas from other bubbles by coalescence, at the same concentration \( C_b \)

(3) by addition of gas from the particulate phase at the concentration \( C_e \).

A mole balance is made to examine the change of moles of tracer in the bubble between time \( t \) and \( t + \Delta t \)

\[
\left. \frac{V_b C_b}{V_b} \right|_t - \left. \frac{V_b C_b}{V_b} \right|_{t+\Delta t} = - K_{be} \left. V_b (C_b - C_e) \right|_t \Delta t + \left. \frac{-\Delta V_{be}}{b} \right|_t C_b + \Delta V_{be} C_e.
\]

\[
\begin{align*}
(1) \\
(2)
\end{align*}
\]

\( \Delta V_{be} \) is the gas which has entered the bubble from the emulsion. Dividing by \( \Delta t \), taking the limit \( \Delta t \to 0 \), changing the time variable and then rearranging the above equation becomes

\[
\frac{u_b}{b} \frac{dC_b}{dh} = - K_{be} (C_b - C_e) - u_b f(h) (C_b - C_e)
\]
where \( f(h) = \frac{\Delta V_{be}}{\Delta x} \), when \( \Delta x \to 0 \), which is the rate of exchange with height from the bubble to the emulsion. Note that this term could be easily included with the other exchange coefficient and that it is probably negative, i.e. gas should generally flow from the bubble to the emulsion when two bubbles interact. Experimentally, Grace and Venta (1973) and Werther (1976) have found this phenomena, and the opposite i.e. gas flowing from the emulsion to the bubble.

According to Pyle (1970), Toei, Matsuno and Nishitani have suggested the same process but taking into account a specific coalescence. If Pyle's (1972) exposition is faithful, their analysis is dubious. Werther (1973) has done some work in this aspect. As he observes, leakage from one bubble to another through the emulsion must be an important mechanism of mass transfer. The above suggestion does not account for this.
Mixing in the Bubble Phase

The diffusion equation V-1 may be solved by separation of variables. Let \( C(r, z) = R(r)Z(z) \). Then the differential equation becomes

\[
\frac{r^2}{dr} \frac{d^2 R}{dr^2} + \frac{dR}{dr} + \frac{\lambda^2}{D_r} r^2 R = 0 \tag{AV-1-1}
\]

for the radial part and

\[
\frac{d^2 Z}{dz^2} - \frac{v}{D_z} \frac{dZ}{dz} - \frac{\lambda^2}{D_a} Z = 0 \tag{AV-1-2}
\]

for the axial part \( \lambda^2 \) is the separation constant. The solution to the Bessel equation (1) is

\[
R_i = A_i J_0 \left( j_{k,i} \frac{r}{r_o} \right)
\]

where

\[
J_{k,i}(j_{k,i}) = 0 \text{ for } i = 0, 1, 2, \ldots
\]

and \( k = 0 \) if the bubble wall is at constant concentration \( C_w \) or \( k=1 \) if the bubble wall is impermeable. The finiteness of \( C \) at \( r = 0 \) has been assumed. The separation parameter has become \( \lambda = j_{k,i} D_t^{1/2}/r_o \).

\( j_{0,0} \) is undefined.

As it is assumed that \( vL/D_a \gg 1 \), the second order term in Eqn. (2) may be neglected with respect to the first order term.
\[
\frac{dZ}{dz} + \frac{\lambda^2}{v} Z = 0
\]

Its solution is
\[
Z = A e^{-\frac{\lambda}{v} z}
\]

Replacing \( \lambda \) for the determined value and combining the separated solutions, the complete solution is obtained.

\[
C = \sum_{l=0}^{\infty} \alpha_{k,i} J_0(j_{k,i} \frac{r}{r_0}) \exp \left( -\frac{j_{k,i}^2 Dr}{v r_0^2} \right) \tag{A2-1-3}
\]

for \( k = 0,1 \), depending whether the walls of the cylinder are assumed at a fixed concentration or impermeable, respectively, and

\[
\alpha_{k,i} = \frac{\int_{r_o}^{r_o} C(r,o) r J_0(j_{k,i} \frac{r}{r_0}) \, dr}{\int_{r_o}^{r_o} r J_0^2(j_{k,i} \frac{r}{r_0}) \, dr}
\]

for \( K = 0,1; i = 0,1,2\ldots \) except

\[
\alpha_{o,o} = C_w
\]
Slow Bubbling Bed Dispersion Model.

AV-2.1 **Two dimensional bed.** The two dimensional bed is assumed to consist of a grid of stationary bubbles distributed in horizontal layers. The bubbles in the same layers are 2 \( \frac{1}{4} \) apart and the layers themselves are \( \frac{1}{2} \) apart. Their relative positions are as shown in fig. (1). There is an equal probability of \( \xi \) for the gas to go from one bubble to one of the two closest to it in the layer immediately above and the rest goes through the bubble in the second layer directly above.

This model can then be looked at as a one dimensional random walk with probability of no motion, which will be solved by the method suggested by Feller in his "Introduction to Probability" (1968). Consider a particle of tracer gas at a horizontal layer \( h \) layers above the one where the tracer gas was originated, and \( m \) bubble positions to the side. The probability of the gas reaching this point is equal to the tracer concentration at this point divided by the concentration at the original bubble.

The gas in a bubble has to go upwards. The step it may take is either to the bubble at its left or to the one at its right in the layer immediately above or to the bubble immediately above two layers up. One of them will indicate whether the gas has gone to the left (-1) or the right (1) or gone directly up (0) and the other will indicate the number of layers moved within the step. So, for the permissible steps
Slow Bubble in a Two Dimensional Array

Figure A-2-1
indicated above, the numbers would be (-1, 1), (1,1) and (0, 2) respectively. The probability for these steps to take place are \( \xi \), \( \xi \) and \( (1- \xi) \).

The probability of arriving from the bubble at the origin to one at the position \((m,h)\) (m bubbles to the right, h horizontal layers above) is proportional to the number of paths connecting both bubbles. Following Feller's method, an array is formed which indicated the path followed by the particle. The element at the j'th position of the array is either (-1) (a move to the left), (1) (a move to the right) or 0 (no move) from the \((j-1)\)th horizontal layer. The 0's must come in pairs as this is the case where a layer is by-passed. So, a path to \((m,h)\) could be represented by an array of \( h \) elements, the sum of the elements adding up to \( m \). For example the array of \( h \) terms

\[
(1, 0, 0, 1, -1, 1, 0, 0, 0, 0, -1, \ldots)
\]

would lead to the point \((m,h)\) if

\[
m = 1 + 0 + 0 + 1 - 1 + 1 + 0 + 0 + 0 - 1 + \ldots
\]

If an array with 2k zeros is to correspond to a path to \((m,h)\), it must have \( p(1)'s \) and \( q(-1)'s \) such that

\[
p + q = h - 2k
\]

\[
p - q = m
\]

Then, the number of arrays with 2k zeros, \( p(1)'s \) and \( q(-1)'s \) is:

\[
\left( \begin{array}{c}
\text{number of different ways} \\
\text{K-pairs of (0) can be positioned in the h-dimensional array}
\end{array} \right) \times \left( \begin{array}{c}
\text{number of different ways p(1)'s may be placed in an (h-2K)-dimensional array}
\end{array} \right) =
\]
\[
= \binom{h - K}{2K} \times \binom{h - 2K}{p}
\]

The probability of taking any one of these paths is

\[
\xi^{p+q} (1 - 2\xi)^{2k}
\]

So the probability of taking any path to \((m, h)\) with \(k\) by-passes is

\[
P_{m, h} = \binom{h - k}{k} \left( \frac{h - 2k}{2} + \frac{|m|}{h - 2k} \right) \xi^{h-2k} (1 - 2\xi)^{2k}
\]

Note that \(h + m\) must be even as only in these positions is there a bubble.

For positions where \(h + m\) is odd, an imaginary bubble

may be placed where the gas comes from below and goes directly above.

In this case

\[
P_{m, h} = (1 - 2\xi)P_{m, h-1}
\]

AI-2.2 Three Dimensional Bed. The arguments for the three dimensional bed

are very similar. In this case the random walk is a two-dimensional one

with a probability of zero motion. The array representing the path of a

fluid particle from the bubble source now will be \((-1)\) (a move in the \(-x\) direction), \(1\) (a move in the \(x\)-direction), \(-i\) (a move in the \(-y\) direction), \(i\) (a move in the \(y\) direction) or 0 (no move). So a path leading to the

point \((m, n, h)\) from \((0, 0, 0)\) will be presented by an \(h\)-dimensional array

where the sum of the elements is \(m + n\). An example would be

\((0, 0, 1, -1, 0, 0, i, l, -i, l, -l, -l, -i,...)\)

which is \(h\) long and
\[ m + i \ n = 0 + 0 + 1 - 1 + 0 + 0 + i + 1 - i + 1 - 1 + 1 - i + \ldots \]

A path arriving at \((m, n, h)\) with \(2k\) zeros, \(r(i)'s\), \(s(-1)'s\) will have \(p(i)'s\) and \(q (-1)'s\) such that

\[
\begin{align*}
r - s &= h \\
p + q &= h - 2k - r - s \\
p - q &= m
\end{align*}
\]

The probability of reaching \((m, n, h)\) with \(k\) and \(r\) given is

\[
\frac{\text{probability of reaching } (m,n,h)}{\text{by any one of these paths}} \times \frac{\text{number of different ways } k \text{ pairs of } 0'\text{s may be placed in the array}}{\text{of } h \text{ elements}} \times \\
\frac{\text{number of different ways } r(i)'s \text{ may be placed in an array of } h-2k \text{ elements}}{\text{number of different ways } s(-1)'s \text{ may be placed in an array of } h-2k-r \text{ elements}} \times \\
\frac{\text{number of different ways } p (1)'s \text{ may be placed in an array of } h-2k-r-s \text{ elements}}{\frac{\binom{h-2k}{h-k} (1-4\xi)^k \binom{h-k}{k}}{\xi}} \times \\
\frac{\binom{h-2k}{h-2k-r} \binom{h-2k-2r}{h-2k-2r+|n|+|m|}}{r-|n|} \times \\
\frac{\binom{h-2k-2r}{h-2k-2r+|n|+|m|}}{r-|n|}
\]

The total probability of reaching \((m, n, h)\) will then be

\[
P_{m,n,h} = \sum_{r=|n|}^{h+|n|-|m|} \sum_{k=0}^{\Sigma^2} \left( \begin{array}{c} h-k \vspace{.1in} \\ r \end{array} \right) \left( \begin{array}{c} h-2k \vspace{.1in} \\ r \end{array} \right) \left( \begin{array}{c} h-2k-r \vspace{.1in} \\ r-|n| \end{array} \right) \times \\
\left( \begin{array}{c} h-2k-2r+|n| \vspace{.1in} \\ h-2k-2r+|n|+|m| \end{array} \right) \left( 1-4\xi \right)^k \xi \vspace{.1in} ; (m+n+h) \text{ even}
\]

\[ k = h-2k \]
Due to the numbering method, \( m + n + h \) must be even for there to be a bubble in the given position. If \( m + n + h \) is odd, a probability may be defined as

\[
P_{m,n,h} = (1 - 4 \xi) P_{m,n,h}^{(m+n+h) \text{ odd}}
\]

The concentration at \((m, n, h)\) due to a bubble at \((0,0,0)\) is

\[
C(m, n, h) = C(0,0,0) P_{m,n,h}
\]
APPENDIX V-3

AV-3.1 Value of the Probability $\xi$ When $l_2 >> d_b$ for Three Dimensional Beds

According to the Davidson and Harrison model, when $l_2 >> d_b$, the probability $\xi$ becomes independent of $l_2$. In practice it is sufficient for $l_2 \approx 2d_b$. Under this condition, the flow through $(3/2)d_b$ above a bubble has reduced its velocity to $u_{mf}$ and has consequently expanded into a cylinder of diameter $\sqrt{3}d_b$. As the flow at this level may be considered uniform, the calculation of $\xi$ is reduced to a geometric problem, which consists of evaluating the fraction of the cylinder which overlaps with the cylinders corresponding to the bubbles receiving the gas from the layer which is immediately above. $\xi$ is a quarter of this fraction.

The problem may be divided into four different cases.

a) If $l_1/d_b > \sqrt{3}$ the cylinders have no overlapping areas. Hence $\xi = 0$, and the bubbles do not aid much in the mixing.

b) If $\frac{\sqrt{3}}{2} < l_1/d_b < \sqrt{3}$ the cylinders do overlap with the one below, but not among themselves, as shown in Fig. 1.3. The area of overlap is

$$A_o = \int_{0}^{\sqrt{3}d_b/2} \arccos \frac{l_1}{\sqrt{3}d_b} r d\theta dr - 2\frac{l_1}{\sqrt{3}d_b} \sqrt{d_b^2 - \frac{l_1^2}{4}}.$$ 

Also $A_o = 3\pi d_b^2 \xi$. (AV-3.1)

From these two eqns. it is found that

$$\xi = \frac{2}{\pi} \arccos \left( \frac{l_1}{\sqrt{3}d_b} \right) - \frac{2}{3\pi} \frac{l_1}{d_b} \sqrt{3 - \left( \frac{l_1}{d_b} \right)^2}$$
Figure AV-3-1a

Figure AV-3-1b

Different Cases of Overlapping Cylinders
c) If \( \frac{\sqrt{3}}{2} < \frac{\ell_{1/d_b}}{} < \frac{\sqrt{3}}{2} \) the cylinders of a same layer will overlap, but will not completely cover the cylinders below them, as shown in Fig. (1b)

\[ A_o = \frac{3\pi d_b^2}{4} - 4 \left[ \frac{\sqrt{3}}{2} \ell \frac{d_b}{d_b} \sin \left( \frac{\pi}{4} - \arccos \frac{\sqrt{2}}{3} \frac{\ell_{1/d_b}}{d_b} \right) \right] - \frac{3}{4} d_b^2 \left( \frac{\pi}{4} - \arccos \frac{\sqrt{2}}{3} \frac{\ell_{1/d_b}}{\pi d_b} \right) \]

The first term in the square brackets represents the area OBCD and the second term the circular section CBD in Fig. (2). Combining this equation with equation (1)

\[ \xi = \frac{1}{2} - \frac{1}{\pi} \arccos \frac{\sqrt{2}}{3} \frac{\ell_{1/d_b}}{d_b} - \frac{2}{\sqrt{3}} \frac{\ell_{1/d_b}}{\pi d_b} \sin \left( \frac{\pi}{4} - \arccos \frac{\sqrt{2}}{3} \frac{\ell_{1/d_b}}{d_b} \right) \]

d) \( \ell / \sqrt{2} < \frac{\ell_{1/d_b}}{} < \sqrt{3} \). In this range there is no by-passing so \( \xi = 0.25 \). Note that if \( \frac{\ell_{1/d_b}}{} \leq \frac{1}{\sqrt{2}} \) the bubbles in the same horizontal layer would geometrically interfere.

AV-3.2 Relationship Between Bed Properties. Consider the prism consisting of a bubble and its eight neighboring bubbles, half on the layer above and half below. The volume of this rectangular prism is \( \delta \frac{\ell_1 \ell_2}{2} \). If every bubble is surrounded by a similar prism, only two prisms will contain any given point. So half of the volume within the prism may be uniquely corresponded to a given bubble. The ratio of the bubble volume to the latter
volume is the volume fraction of bubbles in the bed. Hence

\[ \delta = \frac{\pi d_b^{3/6}}{4 \ell_1^2 \ell_2} \]

Rearranging

\[ \ell_2 = \frac{\pi d_b^3}{24 \ell_1^2 \delta} \]  \hspace{1cm} (AV-3.2)
Calculation of the Variance of the Concentration in a Three-Dimensional Bed

AV-4.1 Variance of the slow bubbling bed model. The variance of the concentration across a horizontal plane may be defined as

\[ \text{Var}(h) = \sum_{m,n} \left( \frac{\varnothing^2}{(m+n)^2} \right) \frac{C(m,n,h)}{C(0,0,0)} \]

where \( m, n \) are the positions of bubbles on a grid, \( h \) horizontal layers above the source, \( C(m, n, h) \) is the concentration at this bubble and \( \varnothing \) is the unit length of the horizontal grid.

As \( \varphi_{m,n,h} = \frac{C(m,n,h)}{C(0,0,0)} \)

and \( \varphi_{m,n,h} = \varphi_{-m,n,h} = \varphi_{m,-n,h} \)

then \( \text{Var}(h) = \sum_{m,n} \left( \varnothing^2 \right) \varphi_{m,n,h} \)

Feller shows that for an ordinary random walk the variance is a linear function of height. This is also true for the continuous case, as will be seen in the next section. However, the variance is not exactly linear for the random walk which has been studied here due to the allowance of by-passing.

It was found by a number of trials that the variance was well approximated by a straight line extrapolated from the variance at \( h = 2 \) and \( h = 4 \). The values of the variance at these heights are
\[ \text{Var}(2) = 4 \xi (1 + 4\xi) \]
\[ \xi \]
\[ \text{Var}(4) = 8 \xi (32 \xi^3 - 8\xi^2 + 2\xi + 1) \]
\[ \xi \]

The straight line that goes through the points \((2, \text{Var}(2))\) and \((4, \text{Var}(4))\) is

\[ \text{Var}(h) = 2 \xi (64 \xi^3 - 16\xi^2 + 4\xi + 1)h \]
\[ \xi \]

AV-4.2 Variance of the continuous model. This model is used to calculate the variance from Hiraki et al.'s (1968) and Rowe and Evans' (1974) results. The differential equation to be solved assumed fluid flowing in plug flow with both radial and axial dispersion coefficients

\[ v \frac{\partial c}{\partial z} = D \frac{\partial^2 c}{a \partial z^2} + \frac{D_r}{r} \frac{\partial}{\partial r} (r \frac{\partial c}{\partial r}) \]

For the simplicity, the medium is considered of infinite extent and the dispersion coefficients equal. As in the slow bubbling bed model a point source is assumed to be at \(r = 0\). The solution was first given by Wilson for a heat transfer problem and later by Towle and Sherwood as a mass transfer problem. The solution is

\[ \frac{C(x,y,z)}{C_a(z)} = z \exp \frac{2v}{D} \left( z - \sqrt{\frac{x^2 + y^2 + z^2}{x^2 + y^2 + z^2}} \right) \]

for \(z \geq 0\) and \(x^2 + y^2 = r\). \(C_a\) is the concentration at the axis. This solution was also given by Klinkenburg et al. and Bischoff and Leven-
The variance of the concentration profile is given by

\[ \text{Var}(z) = \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} (x+y)^2 \frac{C(x,y,z)}{C_a(z)} \, dx \, dy}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{C(z,y,z)}{C_a(z)} \, dx \, dy} \]

The integrals are best solved in cylindrical coordinates \((r, \theta, z)\). After integrating in \(\theta\), the change of variable \(l = \frac{\sqrt{2 \frac{2}{r+z}}}{z}\) is performed and the solution is

\[ \text{Var}(z) = \frac{16 \pi z^2}{2 \text{Pe}^2} (1 + \frac{1}{\text{Pe}}) \]

where \(\text{Pe} = \frac{uz}{D}\)

Simplifying the above expression

\[ \text{Var}(z) = \frac{4D}{u} \left( z + \frac{2D}{u} \right) \]

Note that in general \(\text{Pe} \gg 1\), so it may be simplified to

\[ \text{Var}(z) = \frac{4Dz}{u} \]
Correction of the Nelson and Galloway Theory

The Nelson and Galloway equations were resolved by using the integral heat conduction equation in spherical coordinates

\[- \left( \frac{d}{2} \right)^2 D \frac{\partial C}{\partial r} \bigg|_{r = \frac{d}{2}} = \frac{3}{8} \int_0^d \frac{d}{dr} \left( \frac{d}{2} + \rho \right)^2 \rho C \frac{\partial \rho}{\partial t} \bigg|_{r = \frac{d}{2} + \rho} \tag{AVI-1-1} \]

where \( \rho(t) \) is the penetration depth of the concentration boundary layer.

The penetration depth grows from the particle surface at \( t=0 \) up to the fluid shell thickness \( \delta \) at \( t \geq t_d \) where \( t_d \) is the time required for the penetration thickness to reach the outer shell, i.e. \( \rho(t_d) = \delta \). For \( t > t_d \) (which occurs only if \( t_d > T \), \( T \) being the surface renewal period) then \( \rho = \delta \). An hyperbolic concentration profile is assumed

\[ C = \frac{B_1}{r} + B_2 + B_3 r \tag{AVI-1-2} \]

where the \( B_i \)'s are functions of time and the penetration thickness. The conditions imposed on the profile are

\[ C \left( \frac{d}{2}, t \right) = C_0 \]  \hspace{1cm} \text{if } t \leq t_d \tag{AVI-1-3a} \]

\[ C \left( \frac{d}{2} + \rho, t \right) = \begin{cases} C_0 & \text{if } t \leq t_d \\ C_1(t) & \text{if } t > t_d \end{cases} \tag{AVI-1-3b} \]
\[
\frac{3c}{3r} \bigg|_{r = \frac{d_p}{2} + \rho} = 0 \quad \text{(AVI-1-3c)}
\]

The B's are found from the above conditions. The profile is then substituted into Eqn (1) which for \(0 \leq t \leq t_d\) is transformed into an ordinary differential equation in \(\rho^2\):

\[
\frac{d\rho^2}{dt} = 12 \quad \text{(AVI-1-4)}
\]

which is readily solved by using the boundary condition \(\rho(0) = 0\):

\[
\rho = \sqrt{12t} \quad \text{(AVI-1-5)}
\]

Substitution into the concentration profile and calculation of the time averaged mass transfer from the particle per unit area yield a Sherwood number

\[
Sh_p = 2 + \frac{2 \frac{d_p}{\sqrt{3Dt}}}{2} \quad \text{(AVI-1-6)}
\]

If the surface renewal period \(T\) is less than \(t_d\), then \(t = T\); and from eqn (VI-7)

\[
Sh_p = 2 + 0.69 \frac{Re_p^{1/2}}{Sc^{1/3}} \quad \text{(AVI-1-7)}
\]
This is, coincidentally, Rowe et al's [32] equation for a single sphere. When the surface renewal time $T$ is larger than $t_d$, the solution must be continued. Again the $B_1$'s are found from Eqns. (2) and (3) and substituted into Eqn. (1), which now becomes a differential equation in $C_I$ as $p$ is now fixed ($p = \delta$):

$$\frac{dC_I}{dt} = \frac{-6}{\delta^2 (8 \delta^2 + 11 \delta d_p + 4 d_p^2)} \frac{d^3 (d + \delta) C_I}{d p^3}$$

(AVI-1-8)

with boundary conditions $C_I(t_d) = C_0$.

To calculate the Sherwood number, the mass transfer rate and the particle driving force ($C_o' - C(d_p/2 + \delta, t)$) must be averaged over $0 < t < T$ by use of Eqn. (2). Replacing this in eqn(VI-8) yields

$$Sh_p = \frac{2(d + \delta)(b - c)}{\delta(b - d_p^2 - c)}$$

(AVI-1-9)

where

$$b = 2(4 \delta^2 + 6 \delta d_p + 3 d_p^2)$$

(AVI-1-9a)

$$c = (8 \delta^2 + 11 \delta d_p + 4 d_p^2) \exp\left[\frac{d_p (d + \delta)(12 DT - \delta^2)}{\delta^2 (8 \delta^2 + 11 \delta d_p + 8 d_p^2)}\right]$$

(AVI-1--b)
As \( \frac{\delta}{d_p} = \frac{1 - (1 - \varepsilon)}{2(1 - \varepsilon)^{1/3}} \) and \( T \) is given by Eqn.(VI-7), \( \text{Sh}_p \) is really a function of \( \text{Re}_p \), \( \text{Sc} \), and \( \varepsilon \). This is plotted in Fig. 3.
Alternative Model for Concentration Profiles in a Fixed Bed - I

The solution of Eqn.(VI-15) with boundary condition given by Eqn.(VI-17),

\[ C_1(0) = C_2(0) \]  \hspace{1cm} (AVI-2-1a)

and no mass flux at \( z = 0 \),

\[ \bar{u} C_1(0) = \sum_{j=1}^{2} \frac{\alpha_j D_{e,j} \frac{\partial C_j}{\partial z}}{\partial z \mid_{z=0}} \]  \hspace{1cm} (AVI-2-1b)

is straightforward but lengthy. The general solution of the ordinary differential equation is found for both regions and after satisfying the boundary conditions the exit concentration is given by

\[ \frac{C_e}{C_s} = 1 - 4 \sum_{j=1}^{2} \alpha_j \frac{Re_p \frac{d}{d_p}}{Re_d \frac{d}{d_j}} \left[ \xi_j \exp(-2 \beta_j) - \kappa_j \exp(\gamma_j) \right] \]  \hspace{1cm} (AVI-2-2a)

where

\[ \xi_j = \frac{1 - \eta_{3-j}}{\Delta} \]  \hspace{1cm} (AVI-2-2b)

\[ \kappa_j = \frac{\eta_j (1 - \eta_{3-j})}{\Delta} \]  \hspace{1cm} (AVI-2-2c)
\[ \eta_j = \frac{1 + \alpha_j}{1 - \alpha_j} \exp(\beta_j) \]  
(AVI-2-2d)

\[ \lambda_j = \frac{a_j \Re \bar{p}_j \bar{d}_p}{2 \Re \bar{p}_j \bar{d}_p} (1 + \alpha_j) \]  
(AVI-2-2e)

\[ \varphi_j = \frac{a_j \Re \bar{p}_j \bar{d}_p}{2 \Re \bar{p}_j \bar{d}_p} (1 - \alpha_j) \]  
(AVI-2-2f)

\[ \Delta = -1 - \eta_1 \eta_2 + \sum_{j=1}^{2} [(\lambda_j - \eta_j \varphi_j)(1 - \eta_{3-j}) + \eta_j] \]  
(AVI-2-2g)

and \( \alpha_j, \beta_j \) and \( \gamma_j \) are given in Eqn. (VI-18b-d)
Alternative Model for the Concentration Profile in a Fixed Bed-II

A third model of the mass transfer in a packed bed was considered. The concentration profile across the bed was assumed constant, i.e. the effective radial diffusion coefficient was taken to be infinite (or Pe=0). A mass balance applied to an element covering the whole bed cross section and (dz) high is sufficient to define the required differential equation:

$$\bar{\varepsilon} \frac{\bar{D}_e}{d} \frac{d^2 C}{dz^2} - \bar{u} \frac{dC}{dz} - \frac{6k(1-\varepsilon)}{d_p} (C - C_s) = 0$$

with boundary conditions:

$$\bar{\varepsilon} \frac{\bar{D}_e}{d} \frac{dC}{dz} \bigg|_{z=0} = \bar{u} C \bigg|_{z=0}$$

and

$$\frac{dC}{dz} \bigg|_{z=L} = 0$$

where

$$\bar{D}_e = (a_1 D_{e,1} \varepsilon_1 + a_2 D_{e,2} \varepsilon_2) / \bar{\varepsilon}$$

and

$$\bar{k} = [a_1 k_1 (1-\varepsilon_1)/d_{p,1} + a_2 k_2 (1-\varepsilon_2)/d_{p,2}] d_p / (1-\varepsilon)$$

The solution to this system of equations is:

$$\frac{C_e}{C_s} = 1 - \frac{4\alpha \exp[-\frac{Pe \frac{L}{d_p}}{2d_p} (1-\alpha)]}{(\alpha+1)^2 - (\alpha-1)^2 \exp(-\frac{Pe \frac{L}{d_p}}{d_p} \alpha)} \quad (AVI-3-1)$$
where

\[
\alpha = 1 + 24 \frac{k \frac{D_e}{D_e} (1-\varepsilon)}{\sqrt{\frac{\bar{u}^2}{\bar{u}^2 d_p}}} \quad (AVI-3-2)
\]

and

\[
Pe = \frac{\bar{u} d_p}{D_e}.
\]

The results of this model tend to give exit concentrations which, when reprocessed to find the derived Sherwood number, give values which are quite similar to the actual particle Sherwood number. However, this model should be considered unrealistic as lateral concentration gradients must be of less importance than the axial gradients.
The Computer Program

The program used for calculation of the derived Sherwood number as a function of the average particle Reynolds number is presented here. Three models for mass transfer were used to calculate the exit concentration. These are described by equations VI-18 and in Appendices (2) and (3). The effects of the errors indicated in chapter VI may be analyzed separately or combined by proper choice of bed parameters, effective Peclet numbers and the flags. If wrong effective diffusion equations are to be used, the corresponding Peclet number (divided by 2) should be defined by the user in a subroutine PECLE, as shown in the listing (there B represents the inverse of the assumed Peclet number). M4 helps by indicating which set of data does a set of data corresponding to a flow chart is found in fig 1. The computer program uses the plotting routine QPICTR, which is found in the program library of the Civil and Mechanical Engineering Joint Computer Facility at MIT. The input-output codes for READ and WRITE statements are also those used by these computers. In all other respects, the program is written in general FORTRAN IV. Meaning of the relevant symbols are given at the beginning of the listing as comment statements.
--- Initial \( \frac{b_p}{D} \), number of decades of \( b_p \) to be spanned
--- Coefficients of \( \eta \)
--- \( a_1x_1, a_2x_2/\eta, x_3, x_4, \frac{\eta}{D} \)
--- Options

Compute \( x_2 \frac{x_1}{\eta} \)

Increment \( \frac{b_p}{D} \)
Compute \( b_{p,1} \) and \( b_{p,2} \)
Compute \( \eta_1, \eta_2, \zeta, \beta \)
\( \delta_{h1}, \delta_{h2}, \) and \( \delta_h \)
\( a_1 \) and \( a_2 \) and \( \gamma \)

Compute \( (1-C_z/C_v) \) by model 1 - eqn(34-18)
model 2 - eqn(34-17-1)
model 3 - eqn(34-1-1)

Adjust for error in measurement ( \( \sigma \) ) by eqn (34-23) in all three models

(1-C_z/C_v)
Calculation of derived \( \delta_h \) from average flow and bed properties

Optional plots and table of derived and theoretical Sherwood numbers for models 1-3 as calculated from above assumptions

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Fig AVI-4-1: Flow Chart of the Computer Program
This program is used to certain derived Sherwood (or Nusselt) numbers from three different models of a packed bed.

Author: F. M. JCCS

DIMENSION SH3(3,46),PE2(4,46),D(3),SH4(3),H1(5,46),H2(5,46),
H3(5,46)
ESF(3,3),XSCI(4)
DIMENSION DFN(2)

M3= NUMBER /OF DIFFERENT SETS CF DATA TO BE CONSIDERED
N=(MAXIMUM REYNOLDS NUMBER TO BE CONSIDERED)/RFO
RFO=MINIMUM REYNOLDS NUMBER TO BE CONSIDERED
XIAM= CONSTANT IN DE IGNY'S(OR VCTUPRA ET AL.'S) PECLET EXPRESSION
CI = CONSTANT IN DE IGNY'S(OR VCTUPRA ET AL.'S) PECLET EXPRESSION
DP= PARTICLE DIAMETER(IN MM.) FOR VCTUPRA'S PECLET EXPRESSION
AE1,AE2= FRACTION CF CROSS SECTION CF RED IN REGION ONE AND TWO
SC= SCHMIDT OR FRANDTI NUMBER
EPS(J) V VCDAGE IN REGION J (J=3 IS AVERAGE VOLTAGE).
XI=(HEIGHT OF RED)/(AVERAGE PARTICLE DIAMETER)
XSI= FRACTION CF PARTICLES IN THE RED WHICH ARE ACTIVE
D(J)= PARTICLE DIAMETER IN REGION J/(AVERAGE PARTICLE DIAMETER)
SIGMA= STANDARD DEVIATION OF GAUSSIAN OF MFASHEEFD CONCENTRATION AS PERCENTAGE OF SATURATION CONCENTRATION

THE FOLLOWING QUESTIONS SHOULD BE ANSWERED BY YES(=1) OR NO(=0)

IDC---IS THIS A MASS TRANSFER--AND NOT HEAT TRANSFER--PROBLEM?
IPLUG---SHOULD MODEL NEGLECTING AXIAL CONFUSION BE USED?
IRAT---SHOULD MODEL USING "REAL" AXIAL DIFFUSION BE USED?
IAAC---SHOULD MODEL WITH AN "ASSUMED" AXIAL DIFFUSION BE USED?
IPICT---SHOULD PLOTS BE MADE?
IWRT---SHOULD TABLE OF DERIVED SH(WH) BE AVERAGE OF PRINTED?
IWRTC---SHOULD LIST OF CONC_REAL A
IWRTC---SHOULD LIST OF CONC=1-C(EXIT)/C(SAT) (REAL AND ASSUMED--IF SIGMA/=0) BE PRINTED?
ITY---WILL PLOTS BE SCALED BY THE USER?

READ(8,1002) M3
D(3)=1.
DO 700 M4=1,M3
READ(8,1003) N,REC,YLAB,CI,DP
READ(8,1000) AE1,SC,FPS(1),FPS(2),XL,XST,D(2),SIGMA
READ(8,1004) IDC,IPLUG,IRAT,IAAD,IPICT,IWRT,IWRTC,ITY

OPTION USE FOR SCALING PLOTS (IF ITY=1)

XSCL(1)=MIN. RE AVERAGE
YSCL(2)=MAX. RE AVERAGE
XSCL(3)=MIN. DERIVED SH
C XSCI(4)=MAX. DERIVED SH
C
IF (IX.NE.1) GC TO 15
READ (8,10C1) (XSCI(J),J=1,4)
IX=28
GC TO 16
15 IX=32
16 CONTINUE

C
C
C SOME PROPERTIES OF THE PED ARE EVALUATED
C
C
AE2=1.-AE1
EPS(3)=AE1*EPS(1)+AE2*EPS(2)
D(1)=(AE1*(1.-EPS(1))/((1.-EPS(3))-AE2*(1.-EPS(2)))/D(2)*3)**0.333333
PUT AE1,EFS,SC,XSI,D, SIGMA,XL

C
C AVERAGE RE(3) IS DEFINED
C
C
DC 500 K1=1,N
STEP=10.**(K1-1)*RFO
DO 400 L=1,10
IF (K1.LT.N.AND.I.EQ.10) GC TO 400
YI=L
RE(3)=YI*STEP
.
C
C ERGUN EQUATION IS SOLVED FOR RE(1) AND RE(2)
C
C
A4=42.85714?86
A8=85.71428571
A=(AE1/AE2)**2-(1.-EPS(1))/((1.-EPS(2))*(EPS(2)/EPS(1)**3*D(2)/
/D(1)
  R=2.*AE1/AE2*(RF(3)/AF2+M4*(1.-EFS(3))/D(2))*F(1)+
  *(1.-EFS(2))*(D(2)*F(2))/F(1))**2/(1.-EFS(3))*(EFS(2)/EFS(1))**2*
  A8*R1=1.*EFS(3)/EFS(4)*(AF2*D(3)+(AF2*D(3)))+(AF2*D(3))**2)*D(1)**2
  D1=R**2-2.*A*C
  IF(D1.LT.0.) GC TO 30
  R1=(R+SOFT(D1))/(2.*A)
  D1=(R-SOFT(D1))/(2.*A)
  IF(R1.LT.0. AND R2.LT.0.) GC TO 30
  IF(R2.LT.0.) GC TO 25
  IF (R1.LT.0.) GC TO 26
  R21=(RE(3)-AF1/D(1)*R1)*D(2)/AF2
  R22=(RE(3)-AF1/F(1)*F2)*D(2)/AF2
  IF(R21.LT.0.) GC TO 21
  IF(R22.LT.0.) GC TO 22
  WRITE(5,2000)
2000 FORMAT(' TWO PAIRS OF RF')
  GC TO 500
22  RF(1)=R1
  RF(2)=R21
  GC TO 35
21  IF(R22.LT.0.) GC TO 23
  RF(1)=R2
  RF(2)=R22
  GC TO 35
23  WRITE(5,2001)
2001 FORMAT(' NO POSITIVE RF2')
  GC TO 500
26  RF(1)=R1
  RF(2)=(RE(3)-RE(1)*AF1)/AF2
  IF(RF(2).GT.0.) GC TO 35
  WRITE(5,2002)
2002 FORMAT(' NO POSITIVE RF COMBINATION')
GO TO 500
26   RE(1)=R2
     RE(2)=(RE(3)-RE(1)*AE1)/AF2
     IF(RE(2)*GE.0.* ) GO TO 35
     WRITE(5,2002)
     GC TO 500
30   WRITE(5,2C03)
2003 FORMAT(' NO POSITIVE RE1')
     GC TO 500
35   DC 50  J=1,3
     IF(IDC.NE.1 ) GC TO 40

(PECLET NUMBER)/2 FOR HEAT OR MASS TRANSFER IS EVALUATED
B3=0.7*EPS(J)/(RE(J)*SC)+XLM /(1.+CL*EPS(J)/(RF(J)*SC))
     GC TO 45
40   XLM1=XLM***(0.28-0.757*ALG10(EPS(J))-0.057*ALG10(XLM))
     B3=XLM1/(RF(J)*SC)+14.5/(DP*(1.+CL/(RF(J)*SC))
45   PF(J)=1./(2.*B3)

THE RANZ EQUATION FOR A SHERWOOD NUMBER
SH2=(2.+0.6*SCRT(RF(J)/EPS(J))*SC**(C.3333))
SH4(J)=SH2

THE FOLLOWING SECTION DEFINES EXIT CONCENTRATIONS FOR THE THREE DIFFERENT MODELS
SH(J)=12.*SH2*PE(J)*(1.-EPS(J))*YSI/(RF(J)*SC)
IF(J, EQ, 3) GO TO 50
ALF(J) = SQRT(1, *SH(J)/PE(J)*P2)
R(J) = EXP(-PE(J)*ALF(J)*XL/D(J))
R(J) = R(J)*P2
50 CONTINUE
DO 60 J = 1, 2
       
FIRST MODEL

DEN(J) = (((1 + ALF(J)*F(J))*P2*(1 + ALF(J)*F(J)))*P2)*R(J)
60 CONTINUE
CCNG(1) = 4, (*AE1*RE(1)/D(1)*EXP(PE(1)*XL/D(1))*R(1)*ALF(1)/DEN(1) +
+AE2*RE(2)/D(2)*EXP(PE(2)*XL/D(2))*R(2)*ALF(2)/DEN(2))
       
SECOND MODEL

G1 = ((1 + ALF(1))/ALF(1)/E(1)*P2
G2 = ((1 + ALF(2))/ALF(2)/E(2)*P2
D1 = AE1*RE(1)/RE(1)*(1 + ALF(1))/D(1)*P2
D2 = AE2*RE(2)/RE(2)*(1 + ALF(2))/D(2)*P2
D3 = AE1*RE(1)/RE(1)*(1 + ALF(1))/D(1)*P2
D4 = AE2*RE(2)/RE(2)*(1 + ALF(2))/D(2)*P2
DEN = (D1+G1+D2)*(1, =G2)+(D3+D4)*G1
A1 = (1, =G2)/DEN
A1 = G1*(1, =G2)/DEN
A2 = (1, =G1)/DEN
A2 = G2*(1, =G1)/DEN
CCNG(2) = (*AE1*RE(1)/D(1)*EXP(RE(1)*XL/D(1))*PE1 +
+AE2*RE(2)/D(2)*EXP(RE(2)*XL/D(2)))*R(3)
THIRD MODEL

\[ \begin{align*}
PE_1 &= PE(1) * PE(2) * (RE(2) * AE2 + AF1 * PE(1)) / (RE(1) * PF(2) * AE1 + AE2 * RE(2) * PE(1)) \\
SH_1 &= (SH(1) * PE(2) * RE(1) / R(1) ** 2 + AE2 / AF1 * SH(2) * PF(1) * RF(2) / D(2) ** 2) \\
& \quad / (PF(2) * RE(1) + AE2 / AE1 * IE(1) * RF(2)) \\
ALF(3) &= SQRT(1. + SH_1 / PF(1) ** 2) \\
E(3) &= EXP(-PE_1 * ALF(3) * XL) \\
CCNC(3) &= 4. * E(3) * EXP(PF1 * X1) * ALF(3) / (((1. - ALF(3)) * F(3)) ** 2 - (1. + ALF(3)) ** 2) \\
\end{align*} \]

IF(IWRTC.EQ.1) PUT RE, CONC
DC 200 J = 1, 3

CONCENTRATIONS ARE ADJUSTED DUE TO MEASUREMENT ERRORS

IF(SIGMA.IE.0.) GO TO 110
A = 0.797884561 * SIGMA * EXP(-0.5 * (CONC(J) / SIGMA) ** 2) / (1. - ERF(CCNC(J) * 0.707106781 / SIGMA))
CONC(J) = CCNC(J) - A
CONTINUE
IF(IWRTC.EQ.1) PUT CONC
CONTINUE
K = 9 * (X1 - 1) + 1
DC 210 J = 1, 3
PF2(J, K) = FE(J) * 2.
SH3(J, K) = SH4(J)
IF(IPLUG.NE.1) GO TO 105
DERIVED SH(NU) WHEN AXIAL DIFFUSION IS NEGLECTED

FSH(1,J) = ALOG(-CONC(J))*RF(3)*SC/(XL* 6.*(1.-FPS(3)))*(-1.)/XSI
H1(J,K) = ESH(1,J)

105 IF(IRAD.NE.1) GO TO 115

DERIVED SH(NU) WHEN ASSUMED AXIAL DIFFUSION IS USED

PI = PE(3)
CALL NEWTON(PI,XL,CONC,ALFA,M,J)
IF(M .GT. 20) GC TO 100
PUT ALFA,M

M = 20 IMPLIES THAT NEWTON DID NOT SATISFY THE CONVERGENCE CRITERION.
CONVERGENCE SHOULD BE GOOD, EVEN SO. THIS IS JUST A WARNING.

100 CONTINUE
FSH(2,J) = (ALFA**2-1.)*FF(3)*RE(3)*SC/(12.*(1.-FPS(3))*YST)
H2(J,K) = ESH(2,J)
115 IF (IAAD.NE.1) GO TO 150

DERIVED SH(NU) WHEN ASSUMED AXIAL DIFFUSION IS USED

PI = PECLIE(M4,FF,EPS,SC)
CALL NEWTON(PI,XL,CONC,ALFA,M,J)
IF(M.EQ.20) PUT AIFA,M
PE2(4,K)=FI*2.
ESH(3, J)=(ALFA**2-1.)*TT
*RE(3)*SC/(12.*(1.-EPS(3))*XSI)
H3(J,K)=ESH(3,J)
150 CONTINUE
210 CONTINUE

DERIVED SH(NU) ARE PLACED IN A MATRIX WITH RE AVERAGE

H1(4,K)=SH2
H2(4,K)=SH2
H3(4,K)=SH2
H1(5,K)=RE(3)
H2(5,K)=RE(3)
H3(5,K)=RE(3)
400 CONTINUE
500 CONTINUE

PLOTS AN TABLES ARE MADE FOR DIFFERENT ASSUMPTIONS ON THE DERIVED SH(NU)

IF(IPLUG.NE.1) GO TO 510
IF(IWRT.NE.1) GC TO 506
WRITR(5,2005)
DC 505 J=1,K
WRITR(5,2004) (H1(J3,J),J3=1,5)
505 CONTINUE
506 IF(IPL T.NE.1) GO TO 510
CALL QPICTR(H1,5,46,CY(1,2,3,4),QX(5),CISCL(IX),QXSCL(XSCL))
PAUSE
510 IF(IRAD.NE.1) GO TO 520
   IF(IWRT.NE.1) GO TO 516
   WRITE(5,2006)
   DC 515 J=1,K
   WRITE(5,2004)(H2(J3,J),J3=1,5)
515 CONTINUE
516 IF(IPILOT.NE.1) GC TO 520
   CALL CPICTR(H2,5,46,QY(1,2,3,4),QX(5),QISCL(IX),QXSC1(XSCL))
   PAUSE
520 IF(IAAD.NE.1) GO TO 550
   IF(IWRT.NE.1) GO TO 526
   WRITE(5,2007)
   DC 525 J=1,K
   WRITE(5,2004) (H3(J3,J),J3=1,5)
525 CONTINUE
526 IF(IPILOT.NE.1) GO TO 550
   CALL CPICTR(H3,5,46,QY(1,2,3,4),QX(5),QTRCL(IX),QXSC1(XSCL))
   PAUSE
550 CONTINUE

C
C ACTUAL AND AVERAGE SH(NU) AND PECLET NUMBER ARE PRINTED
C
C IF(IWRT.NE.1) GC .TC 70C
   WRITE(5,2008)
   DO 560 J=1,K
   WRITE(5,2009) (SH3(J3,J),J3=1,3),(PF2(J4,J),J4=1,4)
560 CONTINUE
700 CONTINUE
2004 FORMAT(5X,5F24.5)
2005 FORMAT(/,' DERIVED SH WHEN AXIAL DIFF IS NEGLECTED','/)

SUBROUTINE NEWTON(PF,XI,CCNC,AIFA,K,J)

THIS SUBROUTINE CALCULATES THE DERIVED SH(NU) FROM AVERAGED VALUES
AND SOME GIVEN AVERAGE PF, BY NEWTON'S METHOD

DIMENSION CONC(3)
FRR=10.***(-5)
AIFA=SQR(1.+(1./(PF*XI)+6.)/(PF*XI))/2.
CC=CCNC(J)*10.***10
CC=ALG(1.-CON/4.)-10.*ALG(10.)

CRUDE APPROXIMATION

DO 5 K=1,20
G=CO+2.*ALG(1.+AIFA)-ALG(AIFA)-PF*XI*(1.-AIFA)
G1=2.*/(1.+AIFA)-1./AIFA+PF*XI
AIFA=AIFA-G/G1
IF(ABS(G).LE.FRR.AND.APS(G/G1).LE.FRR) GO TO 6
5 CONTINUE
6 CONTINUE

SOLUTION

DC 10 K=1,20
C=CONC(J)*((1.+AIFA**2)*SINH(PF*XI*AIFA)+
+2.*AIFA*COSH(PF*XI*AIFA)))+2.*AIFA*EXP(PF*XI)
F1 = CONC(J)*(1. + ALFA)**2 * PF*XL + 2.)*COSH(PF*XL*ALFA) + 
     2.*ALFA*(1.+PF*XL)*SINH(PF*XL*ALFA) + 2.*EXP(PF*XL).
ALFA = ALFA - F/F1
IF (ABS(F/(2.*ALFA*EXP(PF*XL))).LT.FRR .AND. ABS(F/F1).LT.FRR)
  RETURN
10  CONTINUE
RETURN
END

PROGRAM Newton HAS NO ERRORS
FUNCTION FECLE(M4,RE,EPS,SC)

THE ASSUMED FECLET NUMBER/2 SHOULD BE INCLUDED HERE. IN ORDER TO IDENTIFY THEM, THE NUMBER OF CARD SET(M4) IS USED.

DIMENSION RE(3),EPS(3)

EXAMPLE

IF (M4.NE.1) GO TO 10
B=2.
PECLE=1./(2.*B)
GO TO 20
10 CONTINUE
B=C.*5
PECLE=1./(2.*B)
20 CONTINUE
RETURN
END

PROGRAM FECLE HAS NO ERRORS
The Computer Program

GEOM is a subprogram which, together with NEWTON and MAXNEW, define \( m, \beta \) and \( \chi \) for a three-dimensional swarm of bubbles as given by Table (4). The input-output variables are:

(a) Input: 
- \( \alpha \) - ratio of bubble to emulsion velocities (L1)
- \( a/b \) - ratio of bubble diameter to horizontal distance between bubbles (L1)

(b) Output: 
- \( \beta \) - recirculation coefficient
- \( \chi \) - normalized recirculation height
- \( m \) - throughput coefficient

Some of the definitions of the variables used in the subprograms are self-evident. Those which are not are listed in Table 1.
# TABLE AVII-1-1

## Meaning of Symbols in the Program

<table>
<thead>
<tr>
<th>Name in Program</th>
<th>Equivalent Expression</th>
</tr>
</thead>
<tbody>
<tr>
<td>AB</td>
<td>a/b</td>
</tr>
<tr>
<td>ALFA</td>
<td>α</td>
</tr>
<tr>
<td>FI</td>
<td>ψ_o/u_f*a</td>
</tr>
<tr>
<td>QTHRU</td>
<td>m</td>
</tr>
<tr>
<td>VOL</td>
<td>β</td>
</tr>
<tr>
<td>XMAX</td>
<td>X</td>
</tr>
<tr>
<td>YO</td>
<td>γ_o/b</td>
</tr>
<tr>
<td>YC</td>
<td>γ_c/b</td>
</tr>
</tbody>
</table>
SUBROUTINE GEOM

\( a, a/b \)

\( N \)

- Compute \( \alpha, \gamma, y_0/b, \phi_0/(\nu \epsilon) \) from eqns 4, 2, 3(c or d), 6(c or d), respectively
- Compute \( y_c/b \)

\( \beta \)

DEFINES \( \beta \) from Table(4) by use of Simpson's Rule of integration

\( \gamma \)

- Compute \( \gamma \) from eqn(11b)
- Compute \( \chi \) for two dim.array
- Compute \( \chi \) as in Table(4)

\( m \)

- Compute \( m \) from Table(4)

\( \beta, \chi \)

Compute \( \beta, \chi \) for isolated, 2 dim.bubble from Tables(2) and (3) and combine with eqns(13)

\( m \)

Compute \( m \) for isolated, 3 dim.bubble from Table(1)

Fig. AVII-1-1

Flow Chart of the Computer Program
SUBROUTINE GEOM(ALFA, AB, VCI, XMAX, OTHRU)

THIS PROGRAM EVALUATES THE COEFFICIENT OF RECIRCULATION (VOL), THE
NORMALIZED RECIRCULATION HEIGHT (XMAX), AND THE THROUGHFLOW COEFFICIENT
(OTHRU)

AUTHOR: F.M. JOOS

THE INFORMATION NEEDED IS ALFA--THE PURPLE TO EMULSION VELOCITY RATIO
--AND AB--THE RATIO OF THE PURPLE DIAMETER TO HORIZONTAL DISTANCE
BETWEEN PURPLES--

DIMENSION X(5)
PI=3.141592654
IF(AB.LT.0.)GC TO 600

THE FOLLOWING REFERS TO NON-ISOLATED PURPLES

SOME OF THE TYPICAL TERMS ARE DEFINED

ALFAC=(1.-PI*AB/2.*TANH(PI*AB/2.))/(1.+PI*AB/2.*TANH(PI*AB/2.))
T=TANH(PI*AB/2.)*ALFA*TAN(PI*AB/2.)
IF(ALFA.GE.ALFA) GO TO 10

THE HORIZONTAL COORDINATE OF THE STAGNATION POINT OF THE LIMITING STREAMSURFACE IS DEFINED

YO=ACOS(1.-PI*AB*T/(1.-ALFA))/PI
FI=(1.-ALFA)*YO/AB+T*SIN(PI*YO)/(1.-COS(PI*YO))
GO TO 20

10 YO=1.
FI=(1.-ALFA)/AB

THE HORIZONTAL COORDINATE OF THE INTERSECTION OF THE LIMITING STREAMSURFACE AND THE BUBBLE IS DEFINED

20 CALL NEWTCN(AB,YC,ALFA,FI,T)
DY=(YO-YC)/4.

CALCULATION OF THE RECIRCULATION COEFFICIENT (VOL) PROCEEDS

INTEGRATION OF THE AREA BELOW THE LIMITING STREAMSURFACE IS REQUIRED. IT IS DONE BY A FIVE STEP SIMPSON RULE

DC 50 J=1,4
XJ=J
Y=YC+(XJ-1.)*DY
D=T*SIN(PI*Y)/(FI-(1.-ALFA)*Y/AB)+COS(PI*Y)
IF(D.GE.1.) GO TO 25
X(J)=0.
D=1.
GO TO 50

25 CONTINUE
USER=JOOS  201 83763  JOINT COMPUTER FACILITY, MIT

\[ X(j) = \frac{\text{ALOG}(D \times \text{SQR}(D^2 - 1))}{\pi} \]

50 CONTINUE
IF (ALFA.1E.ALFAC) GO TO 80
D = \pi * T * AB / (1. - ALFA) - 1.
\[ X(5) = \frac{\text{ALOG}(D \times \text{SQR}(D^2 - 1))}{\pi} \]
GO TO 90
80 X(5) = 0.
90 \[ A = (X(1) + X(5) + 4. * (X(2) + X(3)) + 2. * X(3) \times GY / 3. -
\[ - (AB^2 \times \text{ATAN}(\text{SQR}(AB^2 - YC^2) / YC) - YC \times \text{SQR}(AB^2 - YC^2)) / 2. \]

C VOL IS CALCULATED BY APPLYING THE CORRECTION FACTOR TO THE ABOVE
RESULT
VOL = A / ((1.3333 - 0.2633 * ALFA) * AB^2)

C CALCULATION OF THE NORMALIZED RECIRCULATION HEIGHT (XMAX) PROCEEDS

C GAMA = \pi * T / ((1. - ALFA) / AB)

C CASE WHERE THE LIMITING STREAMSURFACE HAS A MAXIMUM AT THE MIDPLANE
BETWEEN BUBBLES IS IDENTIFIED
IF (GAMA.1.E.3.) GO TO 400
D = GAMA - 1.

C FOR THE CASE WHEN THE MAXIMUM IS NOT AT THE MIDPLANE...
GO TO 500
400 G = (1. - ALFA) / (\pi * AB)
THE MAX(D) IS FOUND

CALL MAXNEW(YC,YO,C,FI,T,D)

XMAX IS CALCULATED WITH CORRECTION FACTOR

500 XM1= ALOG(D+SQRT(D**2-1.))/(AB*PI)
XMAX=XM1/2.

CALCULATION OF THE THROUGHFLOW COEFFICIENT PROCEEDS

IF (ALFA.GE.ALFAC) GO TO 550

1- FOR NON-ISOLATED TWU DIMENSIONAL CASE

Q=(1.-ALFA)/(PI*AB)*ACCS(1.-PI*AR*T/(1.-ALFA)) + SQRT(2.*(1.-ALFA)*T/(PI*AR)-T**2)
GO TO 560

550 Q=(1.-ALFA)/AR

2- FOR 2 DIMENSIONAL ISOLATED CASE

560 Q1=2.*SQRT(1.-ALFA**2)

3- FOR 3 DIMENSIONAL ISOLATED CASE

Q2=1.889881575*(4.-3.*ALFA**2-ALFA**3)**(1./3.)

FINAL THROUGHFLOW COEFFICIENT
USEH=JOOS 201 B3763 JOINT COMPUTER FACILITY, MIT

QTHRU=0.2*Q2/01
GC TO 700

C
C
C
C IN CASE AB=0 THE TWO DIMENSIONAL ISOLATED BUBBLE PROPERTIES ARE
C USED AND APPROPRIATELY CORRECTED
C
C

600 A1=(2.-ATAN(ALFA/SQRT(1.-ALFA**2))-(?.-ALFA)*SQRT(1.-ALFA**2))*
*(1.+ALFA)/(2.*((1.-ALFA))-(ATAN(ALFA/SQRT(1.-ALFA**2))-
-ALFA*SQURT(1.-ALFA**2))/?,
VO1=A1/((1.3333-0.2633*ALFA)
IF(ALFA.GT.0.618033989) GC TO 660
XM=ALFA
GC TO 670

660 XM=0.30023106*SQRT((1.+ALFA)/(1.-ALFA))
670 XMAX=XM/2.
695 QTHRU=3.*(1.+ALFA/2.)*2.*(1.-ALFA)***(1./3.)
700 IF(VOI.LT.0.) VOI=0.
IF(XMAX.LT.0.) XMAX=0.
IF(QTHRU.LT.0.) QTHRU=0.
RETURN
END

Program GEOM has no errors
SUBROUTINE NEWTON(AB,Y,ALFA,F1,T)

THIS SUBROUTINE FINDS THE COORDINATES OF THE LIMITING STREAM-
SURFACE AND THE BUBBLE

PI=3.141592654

ERROR BAND AND STARTING FCTNT ARE DEFINED

FRH=10.*(-5)
Y1=AB/2.

ITERATION BY NEWTON'S ROOT-FINDING METHOD PROCEEDS

DO 20 J=1,100
X=SQRT(AB**2-Y1**2)
F=(COSH(PI*X/Y1)-COS(PI*Y1))*((1.-ALFA)*Y1/AB-F1)+T*SIN(PI*Y1)
F1=(1.-ALFA)/(AB*(COSH(PI*X/Y1)-COS(PI*Y1)))+PI*((1.-ALFA)*Y1/AB-F1)*(-AB**2/(X*Y1**2))*SINH(PI*X/Y1)+SINH(PI*Y1)+PI*T*SIN(PI*Y1)

THE NEW ESTIMATE AND SOME CHECKS

Y=Y1-F/F1
IF (Y.GT.AB) Y=(Y1+AB)/2.
IF(Y.LT.0.) Y=Y1/2.
IF(ABS(Y-Y1).LT.ERR) GO TO 30
Y1=Y
20 CONTINUE
30 CONTINUE
RETURN
END

PROGRAM NEWTON HAS NO ERRORS
SUBROUTINE MAXNEW(YC,YO,G,FT,T,D)

C THIS SUBPROGRAM FINDS THE MAXIMUM ON THE LIMITING STREAMSURFACE BY
C NEWTON'S ROOTFINDING METHOD APPLIED TO THE DERIVATIVE OF
C
C THE ERROR BAND
C
ERF=10.**(-5)
PI=3.141592654

C LIMITS OF STREAMSURFACE ARE DEFINED
C
RO=YO*PI
RC=YC*PI

C SOME POINTS ON THE LIMITING STREAMSURFACE ARE CHOSEN TO LOCATE THE
C NEIGHBORHOOD OF THE MAXIMUM
C
DC 50 J=1,5
XJ=J*3
R1=RC+(XJ-1.)*(RO-RC)/4.
H=FI-G*R1
F1=T*H*COS(R1)+(T*G-H**2)*SIN(R1)
IF(J.GT.1) GO TO 20

C IF THE SLOPE AT THE BUBBIF-LIMITING STREAMSURFACE INTERSECTION IS
C NEGATIVE, THEN THE INTERSECTION IS THE MAXIMUM
IF(F1.LT.0.) GO TO 110
F0=F1
GO TO 50
20 IF(F1.GT.0.) GO TO 50

A MIDPOINT IS CHOSEN AS THE STARTING POINT

R1=R1-(R0-RC)/8.
GO TO 60
50 CONTINUE
60 CONTINUE

NEWTON ITERATIONS PROCEED

DC 100 M=1,50
H=1.0-G*R1
F1=T*H*CCS(R1)+(T*G-H**2)*SIN(R1)
F2=H*((2.*G-T)*SIN(R1)-H*COS(R1))
R=R1-F1/F2
IF(R.CT.R0) H=(R1+R0)/2.
IF(R.LT.RC) R=(R1+RC)/2.
C=R1-R
R1=R
IF(ARS(C).LT.FR8) GO TO 110
100 CONTINUE

A FINAL CHECK WITH THE RUBBLE-LIMITING STREAMSURFACE INTERSECTION IS MADE

D=T*SIN(R1)/(1.0-G*R1)+CCS(R1)
D1=T*SIN(RC)/(1.0-G*RC)+COS(RC)
IF(D1.GT.D) D=D1
RETURN