3.0 BACKGROUND

3.1 Two-Phase (Gas-Liquid) Flow

Two-phase flow of gas and liquid in vertical vessels has been extensively studied by numerous investigators.[1-28] In most of the studies the liquid is the continuous phase with the gas being dispersed as bubbles. The gas holdup (volume fraction of vessel occupied by gas) in such a system is a key variable in evaluating the performance of a bubble column as a reactor or gas-liquid contactor. Several investigators have developed correlations to predict the variation of gas holdup with independent variables such as gas and liquid superficial velocities, their physical properties, column diameter, and gas distribution mechanisms.

One of the best known correlations was developed by Akita and Yoshida[9] using a dimensional analysis approach:

$$\frac{\varepsilon_g}{(1-\varepsilon_g)^4} = 0.20 \left(\frac{gD^2\rho L}{\sigma}\right)^{1/8} \left(\frac{g\,D^3}{\nu_L}\right)^{1/12} \left(\frac{V_g}{\sqrt{g\,D}}\right)^{1.0} \quad (1)$$

The model excludes the effects of orifice diameter for gas distribution at the inlet to the column, variations in gas density, and liquid superficial velocity, as these variables were found to have insignificant effect on the measured gas holdup.

Hughmark[1] combined data found in the literature with his own to express holdup as a function of the superficial velocities of both phases and liquid physical properties:

$$\varepsilon_g = \frac{V_g}{V_s} \left[\left(\frac{62.4}{\rho_L}\right) \left(\frac{72}{\sigma}\right)\right]^{1/3} \tag{2}$$

V_s is a slip velocity defined as

$$V_s = V_g/\varepsilon_g - V_L/(1-\varepsilon_g) \tag{3}$$

It should be noted that the term in brackets in equation (2) reduces to unity for the air-water system.

Similar correlations have been developed by Hikita and Kikukawa,[24] Calderbank,[25] and Sridhar and Potter,[2] the latter being a modification of the Calderbank equation to account for changes in gas density at elevated pressure.

Much of the work reported in the literature focuses on characterizing the effects of one or several variables on gas holdup. Miller[4] has examined the effects of liquid physical properties and has compared his results to the correlations of Kumar, et al.,[27] Hughmark,[1] Mersmann,[10] Akita and Yoshida,[9] and Anderson and Russell.[26] He concludes that the Anderson-Russell procedure for determining gas holdup is the best for diameters less than 5 inches and that his own procedure is best for diameters greater than 6 inches.

Eissa and Schügerl[22] have also dealt with the variation of gas holdup with changes in liquid viscosity and surface tension. They observed a rapid decrease in gas holdup as viscosity increases between 3 and 11 cp and a more gradual decrease above 11 cp. They conclude that this behavior is caused by an increased rate of bubble coalescence due to higher drag forces on rising bubbles. Concerning surface tension, the authors attribute the observed gradual decrease of gas holdup with increasing surface tension to an increase in terminal bubble velocity. Mendelson[3] presents correlations based on wave theory which predict a square root dependence of terminal bubble velocity on surface tension. Hence, increasing the surface tension results in higher terminal velocities with consequent lower gas holdup.

Freedman, et al.[23] have examined the effect of gas distribution at the inlet to the column on gas holdup and conclude that maldistribution of gas reduces overall holdup. They also conclude that the major source of deviation from behavior predicted by theory is bubble coalescence caused by poor gas distribution. Another possible reason is channeling that may occur as a result of poor distribution of gas at the inlet to the column.

Perhaps the area of poorest agreement in the literature concerns the effect of vessel diameter. Fair, et al.[8] present their data as well as other data reported in the literature and argue that vessel diameter significantly affects holdup to a diameter of approximately 18 inches. Other experimenters, however, claim that the effect of diameter on gas holdup becomes insignificant beyond 4 inches. Botton, et al.[7] have performed an extensive literature review and have concluded that wall effects are significant only up to diameters of 6 to 8 inches.

While there has been ample work at atmospheric pressure, little has been done at elevated pressures more common to industrial applications. The ability to extrapolate correlations developed at low pressure has, therefore, not been thoroughly tested. Sridhar and Potter[2] have examined gas holdup as a function of pressure and stirrer speed in mechanically agitated vessels. They used their results to modify the Calderbank equations predicting interfacial area, mean bubble diameter, and gas holdup to include the effects of pressure. They conclude that gas holdup increases with pressure while mean bubble diameter decreases. They also conclude that Miller's method mentioned above is not adequate at elevated pressures.

3.2 Three-Phase (Gas-Liquid-Solid) Flow

Compared to the amount of literature on two-phase flow, information related to three-phase fluidized beds is much more sparse. However, several important industrial applications using three-phase fluidized beds now exist. These include the catalytic hydrogenation and desulfurization of petroleum, and the liquefaction of coal via the SRC, EDS, or H-Coal processes.

The effectiveness of a three-phase fluidized bed as a reactor relies heavily on the hydrodynamic properties of the bed. The addition of solid particles to a gas-liquid flow system increases the complexity of the system to the point where two-phase flow models become suspect. Solid particles directly influence critical hydrodynamic properties such as gas holdup, liquid dispersion, and backmixing by affecting the behavior of gas bubbles--their formation, coalescence, and breakup. Bubble size, motion, and competing rates of bubble coalescence and disintegration are all related to the degree of solids fluidization or accumulation. The behavior of gas bubbles in three-phase fluidized beds has been the focal point of most experiments reported in the literature to date.[29-43]

Clift and Grace[29] have examined the mechanisms of bubble disintegration in three-phase beds and have defined two types. The first involves the bubble splitting from the rear as a result of the development of an indented wake which, if given enough momentum, can reach the roof of the bubble and cause splitting. The authors claim that this mechanism is only valid as an entrance effect or for pulsed fluidized beds. The second mechanism states that breakup occurs when a knife or curtain of particles descends from the bubble roof. The authors conclude that this behavior, caused by irregularities at the bubble interface due to density variations, is not unlike the mechanism proposed by Taylor[42] to account for bubble formation and is more common than the first mechanism under practical conditions. It should be noted that these mechanisms are more applicable to dense phase fluidized beds and not as applicable to the diluted fluidized bed reactors in the SRC-II process. However, these mechanisms may be pertinent in slurry reactors with a high degree of solids accumulation as was simulated in the solids withdrawal studies described below.

Kim, Baker, and Bergougnou[35,36] have researched many aspects of three-phase fluidized beds. Their work has covered the effects of liquid and gas velocities, and the properties of solids on the relative phase holdups, bed expansion, axial liquid mixing, and bubble characteristics in two- and three-phase beds. Concerning two-phase (gas-liquid) beds, they found that gas holdup increased with increases in both gas and liquid superficial velocity though the effect of the latter was small. Nevertheless, their conclusion concerning liquid velocity contradicts most other work in the literature. For three-phase beds, the authors conclude that the presence of solid particles reduces liquid holdup and increases gas holdup except at the highest gas and liquid rate which they studied. The increase in gas holdup was attributed to a decrease in mean bubble size and rise velocity.

Ying, et al.[28] performed experiments in a comparatively large scale column in support of the dissolver design for the SRC-I process. Concerning gas holdup, they studied the effects of solids particle size and concentration, liquid superficial velocity, and gas superficial velocity. They concluded that gas holdup was independent of liquid velocity and the presence of solid particles at low gas velocity. However, they experienced a decrease in gas holdup for fine particles (<100 mesh) at higher gas superficial velocities (0.368 ft/sec). Except at conditions exceeding those mentioned above, the authors claim that the two-phase correlation of Akita and Yoshida adequately represents their data.

An important aspect of three-phase fluidized beds with fixed solids holdup is the contraction or expansion of the bed upon injection of gas. In three-phase continuous systems such as slurry reactors, the accumulation and concentration gradient of accumulated solids are controlled by the same mechanisms governing expansion or contraction in a bed with constant solids content. Knowledge of this phenomenon is critical to understanding three-phase systems of any kind, so a discussion of these concepts is warranted.

Turner,[43] Stewart and Davidson,[30] and Ostergaard and Theisen[32] have extensively examined bed contraction as it relates to other variables. They propose that the contraction is caused by liquid volume elements moving as wakes behind rising gas bubbles at a velocity higher than the average liquid velocity. The liquid velocity in the rest of the bed thus reduces below the average and the bed contracts. Apparently, the contraction more than compensates for any bed expansion due to higher gas throughput. Ostergaard and Theisen[32] further conclude that bed contraction is greater in beds of small particles than in beds of larger particles. Furthermore, they found that bed contraction increased with increasing bed height.

Dakshinamurty, et al.[31] extended the work of Ostergaard and Theisen to include the effect of particle density, liquid surface tension, and fluid flow rates on bed porosity. Their results for the effect of particle size parallel those of Ostergaard, et al., but they also found that: (1) increasing liquid velocity was followed by a corresponding increase in bed porosity and (2) for a given liquid velocity, a plot of bed porosity versus gas superficial velocity passes through a minimum, the location being dependent upon liquid superficial velocity. Competing effects of bed contraction and expansion are a possible cause for this minimum.

Rigby and Capes[37] used still and moving photography to closely examine the behavior of bubbles, the formation of bubble wakes, and bed contraction in three-phase beds. Their results for bed contraction agree with those of other investigators.[30,31,32] Concerning wakes, the authors conclude that: (1) the proportion of wake associated with a gas bubble passing through a liquid fluidized bed increases with decreasing bubble size and increasing particle size; (2) the bubble wake consists of two portions, a stable portion carried with the bubble and vortices shed by the bubbles; (3) the properties of bubble wakes are the major cause for the contraction of three-phase beds. Agreement between observations made with two different experimental methods supports this claim.

The most important properties affecting the behavior of solids in three-phase systems are particle size and particle density. Kim, et al.[35] propose the existence of two types of three-phase beds. These are termed bubble-coalescing and bubble-disintegrating. The addition of particles smaller than a critical size to a two-phase bed results in an increase in the mean bubble size and a corresponding decrease in gas holdup. Such a bed is termed bubble-coalescing and is usually accompanied by bed contraction. As particle size increases beyond a critical size, the rate of bubble coalescence diminishes and the gas holdup increases because of the presence of smaller bubbles. Thus, the tendency for the bed to contract is greatly reduced. Such a bed is referred to as bubble-disintegrating. The authors estimate, based on the data of Ostergaard, that the critical size may be around 2.5 mm for particles having the density of glass.

Particle size is not the only important variable affecting the behavior of three-phase beds. Particle density, liquid viscosity, liquid and gas velocity, and surface characteristics of the solid-liquid system are all important because they affect the minimum fluidization characteristics of the bed. Calderbank, et al.[25] observed that bubble coalescence was much more pronounced in beds of high viscosity liquids. Kim, et al.[35] conclude that "... solids having a minimum fluidizing velocity in the liquid phase less than

1.28 cm/sec initially contract upon injection of gas into the bed. In contrast, beds of solids having a minimum fluidizing velocity exceeding 1.28 cm/sec expand upon introducing gas."

Other variables that can greatly influence the behavior of a three-phase bed are related to the surface properties of the phases, i.e. surface tension and contact angle. Bhatia, et al.[44] reported that beds of small wettable solids which contracted upon injection of gas reversed behavior and expanded under the same conditions when made nonwettable by being coated with Teflon. It is assumed that the freer motion of the solids increases the rate of bubble breakup, thus increasing gas holdup. Similarly, a reduction in surface tension should increase bed porosity again because the particles are freer to move about due to the decreased surface energy needed to separate the phases.

3.3 Slurry Backmixing in Gas-Liquid-Solid Systems

The extent of backmixing in three-phase flow vessels has been the subject of numerous investigations. Shah, et al.[45] provided an extensive review of these investigations. Generally the approach has been to assume that a simple axial dispersion model adequately describes the mixing patterns in the slurry phase. Estimates of the dispersion coefficient, the parameter that describes mixing in the model, would be obtained from tracer tests. Most of the dispersion data obtained from tracer tests were correlated by an equation of the form:

$$E_z = a\, D_t^{\,b}\, U_{0_G}^{\,c} \qquad \text{a, b, c constants} \tag{4}$$

where

E_z = Dispersion coefficient, cm^2/sec

D_t = Diameter of column, cm

U_{0_G} = Superficial gas velocity, cm/sec

The correlation proposed by Deckwer, et al.(46) is one of the most commonly used correlations for two-phase gas-liquid flow. In this correlation, the constants are: a=2.7, b=1.4, and c=0.3. As can be seen, this correlation does not include physical property effects, if any, on the dispersion coefficient. Cova(47) investigated the effect of fluid properties of both the gas and liquid and concluded that liquid viscosity and surface tension do not have a significant effect on the dispersion coefficient and even the effect of liquid density is only minor, and that it is important only for flow in small tubes.

The effect of suspended particles on axial dispersion in the liquid phase was studied by several investigators: Michelsen and Ostergaard,(48) Ostergaard,(32) Kim et al.,(36) Imafuku et al.,(49) Farkas and Lebland(50) among others. Kato, et al.(51) correlated the longitudinal dispersion coefficient for the slurry phase by the following relationship:

$$E_z = 2.41\, D_t^{1.5} \left[1 + 0.43\, \frac{U_{0_G}^{0.85}}{D_t^{0.43}} \right] \tag{5}$$

Since the assumption that a simple dispersion model can adequately describe the complex flow patterns in the vessel is implicit in the above correlations, a severe problem arises when such an assumption is invalid as when the column is operated outside the bubble flow regime. In these situations a recirculation model based on Davidson's[52] energy balance is more appropriate, since such a model represents the physical structure of flow through the column more closely. In this model, intense recirculation patterns in the liquid phase are visualized. Each circulation pattern is known as a cell and visual observations indicate the presence of these multiple circulation cells in both the axial and radial directions. Joshi and Sharma[53] developed a sound theoretical basis to describe this flow regime which is characterized by intense recirculation patterns in the slurry phase.