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A UNIFIED APPROACH TO THE SCALE-UP OF 'FLUIDIZED' MULTIPHASE REACTORS

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In this paper we stress analogies in the hydrodynamic behaviour of gas-solid fluidized beds, gas-liquid bubble columns and bubble column slurry reactors and suggest a unified approach to scale up of these 'fluidized' multiphase reactors. Using published experimental data it is demonstrated that the analogous hydrodynamic behaviour is not just qualitative but *quantitative* in nature. It is argued that, because of cross-fertilization of concepts and information, appreciation of analogies can be an invaluable tool in scaling up.

Keywords: backmixing; bubble columns; dense phase; dilute phase; fluidized beds; homogeneous regime; heterogeneous regime; hydrodynamics; multi phase reactors; scaleup

INTRODUCTION

There are several books and reviews dealing with the subject of gas–solid fluidized bed reactors and bubble columns[1,2]. Broadly speaking, the hydrodynamic picture of these two important industrial contactors is as described below.

When a gaseous phase is introduced uniformly through the bottom of a packed bed of particles the bed begins to expand for gas velocities exceeding the minimum fluidization velocity U_{mf}. For fine particles, say smaller than 200 μm, the bed expands uniformly; this is the regime of *homogeneous fluidization*. This regime of homogeneous fluidization prevails till a certain velocity is reached at which bubbles are first observed; the velocity at this point, U_{mb}, is usually called the *minimum bubbling velocity*. For the purposes of drawing analogies with gas–liquid systems, we shall denote this velocity as the transition velocity, U_{trans}. The operating gas velocity window between U_{mf} and U_{trans} is usually very narrow and it is usually not possible to operate commercial reactors in a stable manner in this regime. On the other hand in gas–solid beds of large particles, say larger than 1 mm, bubbles appear as soon as the gas velocity exceeds U_{mf} and hence $U_{trans} \approx U_{mf}$. Beyond the gas velocity corresponding to U_{trans} we have the regime of *heterogeneous fluidization*. In the heterogeneous fluidization regime a small portion of the entering gas is used to keep the solids in suspension, while the major portion of the gas flows through the reactor in the form of bubbles. Commercial reactors usually operate in the heterogeneous or bubbling fluidization regime at gas velocities U exceeding 0.1 m s^{-1}, a few orders of magnitude higher than U_{trans}. Under these conditions the bubbles tend to rise up the column very quickly at velocities of the order of 1 m s^{-1}, 'by-passing' the suspended particles. These bubbles tend to churn up the bed causing the solids phase to be thoroughly backmixed. For highly exothermic reactions, such as regeneration of coked catalyst in Fluid Catalytic Cracking (FCC) regenerators, this backmixing characteristic is desirable from the point of view of thermal equilibration of the reactor contents.

An analogous picture emerges if one sparges gas into a column filled with a liquid. The bed of liquid begins to expand as soon as gas is introduced. If we therefore define U_{mf} as the minimum fluidization velocity for a gas–liquid system we see that $U_{mf} = 0$. As the gas velocity is increased, the bed of liquid expands homogeneously and the bed height increases almost linearly with the superficial gas velocity. This regime of operation of a bubble column is called the *homogeneous bubbly flow regime*; this regime is entirely analogous to the regime of *homogeneous fluidization* for a gas–solid system. The bubble size distribution is narrow and a roughly uniform bubble size, in the range 2–7 mm, is found. At a certain gas velocity U_{trans} coalescence of the bubbles takes place to produce the first fast-rising 'large' bubble. The appearance of the first large bubble changes the hydrodynamic picture dramatically. The hydrodynamic picture in a gas–liquid system for velocities exceeding U_{trans} is analogous to the *heterogeneous fluidization regime* for gas–solid systems and is commonly referred to as the *churn-turbulent regime*.

The present paper advocates the use of an unified description of the hydrodynamics of G-S fluid beds, G-L bubble columns and G-L-S slurry bubble columns; see Figure 1.

THE HETEROGENEOUS FLOW REGIME: 'DILUTE' AND 'DENSE' PHASE GAS VOIDAGES

Let us perform a bed collapse experiment in the heterogeneous flow regime of G-S fluid beds and G-L bubble column; in this experiment the gas supply is instantaneously shut off and the height-time information is recorded continuously. Figure 2 shows a typical example of

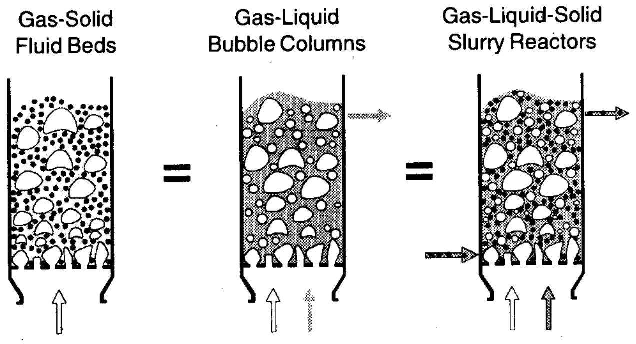

Figure 1. The three types of 'fluidized' multiphase reactors which are considered in this paper.

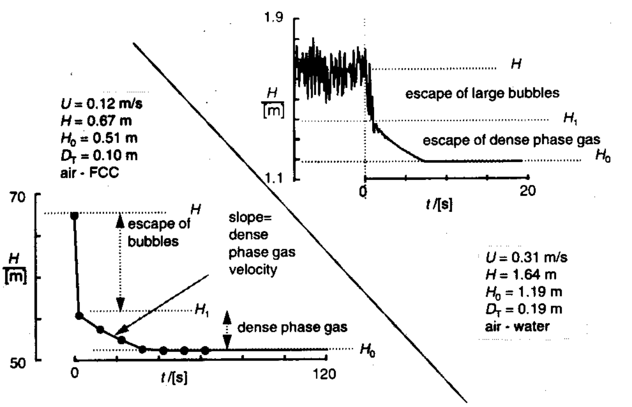

Figure 2. Typical dynamic gas disengagement experiments in G-S and G-L systems.

bed collapse in G-S and G-L systems. The initial sharp decrease in height is due to escape of 'dilute' (= 'bubbles' in G-S fluid beds; =fast-rising 'large' bubbles in G-L bubble columns; this is followed by slow disengagement of the gas entrapped in the 'dense' phase (= 'emulsion' gas in G-S fluid beds; = 'small' bubbles in G-L bubble columns). For G-L bubble columns, typically the 'small' bubbles are 2–5 mm in diameter and their holdup is strongly dependent on the physical properties of the system. From the disengagement curves, the following parameters can be experimentally determined. The total gas voidage, or hold-up,

$$\varepsilon = 1 - \frac{\rho_b}{\rho_p}\frac{H_0}{H}$$

(G-S fluid bed);

$$\varepsilon = 1 - \frac{H_0}{H}$$

(G-L bubble column). The hold-up of the bubbles, or 'dilute' phase, ε_b, is determined from

$$\varepsilon_b = \frac{H - H_1}{H}$$

The gas voidage in the 'dense', or emulsion, phase is

$$\varepsilon_{df} = \frac{H_1 - H_0}{H_1} = \frac{\varepsilon - \varepsilon_b}{(1 - \varepsilon_b)}.$$

The gas voidages for air-FCC and air-water systems are plotted in Figure 3. It can be observed that for both these systems the 'dense' phase gas voidage ε_{df} is practically

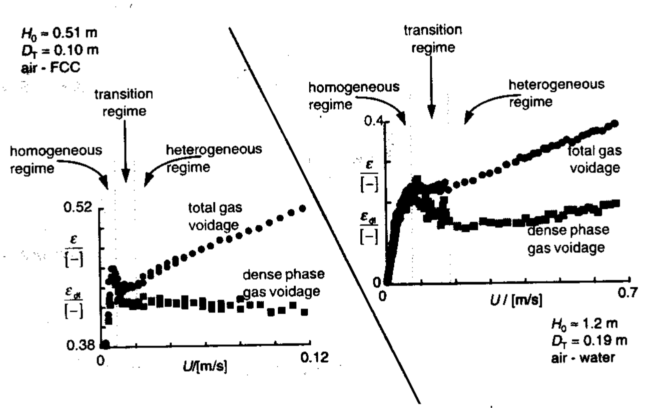

Figure 3. Homogeneous and heterogeneous regimes in G-S fluid beds and G-L bubble columns.

constant, independent of the gas velocity; Ellenberger and Krishna[3] also found ε_{df} to be practically independent of the column diameter. The slope of the second, slowly disengaging, portion of the collapse curves in Figure 2 yields the superficial gas velocity through the dense phase, U_{df}.

A few thousand experimental data were collected for air-FCC, air-polystyrene, air-water, air-tetradecane, air-paraffin oil, and air-5%, 10%, and 20% slurry of paraffin oil in columns of 0.05 m, 0.1 m, 0.19 m and 0.38 m diameter[3–5]. These data when combined with literature data of other research groups[6–12] suggest the following unified hydrodynamic picture for multiphase reactors.

Firstly, we may extend the classic two-phase model for G-S fluid beds[13,14] to G-L bubble columns and G-L-S slurry reactors as shown in Figure 4. The two phases: 'dilute' and 'dense', are to be identified as follows. The 'dilute' phase is to be identified with the solids-free bubbles in a fluid bed or the fast-rising large bubble population in G-L, G-L-S bubble columns. The dilute phase travels up the column virtually in plug flow. The 'dense' phase in a fluid bed consists of the suspension of solids with a gas flow corresponding to U_{df}. For bubble columns the dense phase is to be identified with the liquid phase *together with* the small bubbles which are entrained in the liquid. For G-L-S slurry columns the "dense' phase is identified with the liquid phase along with the solid particles and the entrained 'small' bubbles. In the heterogeneous flow regime, the small bubbles have the backmixing characteristics of the liquid, or slurry, phase. In columns of large diameter the dense phase can be considered to be completely backmixed. The parameter $(U_{df}/\varepsilon_{df})$ represents the 'swarm' velocity of the 'dense'

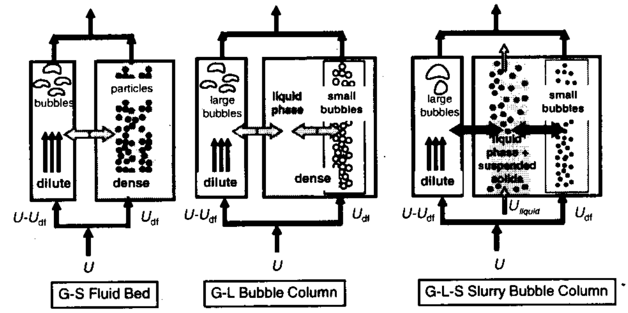

Figure 4. The generalized 'two-phase' model for G-S fluid beds, G-L bubble columns and G-L-S slurry reactors.

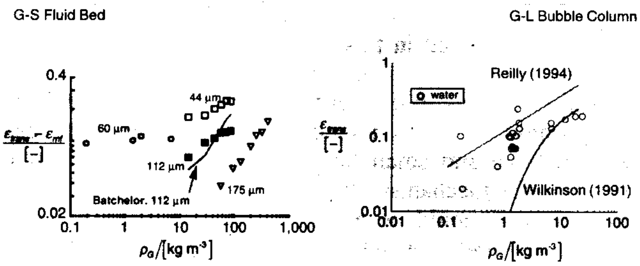

Figure 5. The influence of gas density (equivalent to increased system pressure) on $\varepsilon_{trans} - \varepsilon_{mf}$ is analogous in both G-S and G-L systems. Increased ρ_G (or increase p) increases $\varepsilon_{trans} - \varepsilon_{mf}$.

phase gas. In common with the behaviour of ε_{df}, this 'swarm' velocity (U_{df}/ε_{df} is a scale independent parameter[3]. For G-L and G-L-S systems (U_{df}/ε_{df}) represents the rise velocity of the 'small' bubble fractions, typically in the range of 0.2 m s^1– 0.25 m s^{-1}.

For estimation purposes the gas voidage in the dense phase can be taken to be equal to the gas voidage at the regime transition point $\varepsilon_{df} \approx \varepsilon_{trans}$. The gas voidage at the regime transition point in gas–solid and gas–liquid systems increases significantly with increasing system pressure, or density of the fluidizing gas, as shown in Figure 5; this strong dependence of the regime transition point on gas density can be rationalized using a unified stability analysis[15,16]. A good estimate of the superficial velocity through the dense phase is the regime transition gas velocity, $U_{df} \approx U_{trans}$. The superficial gas velocity in excess of U_{df} rises the column as fast-rising large bubbles. For both fluid beds and bubble columns the regime transition point U_{df} significantly increases with increasing gas density (i.e. pressure).

MODEL FOR 'DILUTE' PHASE GAS HOLD-UP

Figure 6 shows values of the rise velocities of the 'dilute' phase for fluid beds and bubble columns, both of 0.38 m diameter. It is clear that the rise velocities V_b are of similar magnitude, when compared at the same value of $(U - U_{df})$. The 'dilute' phase rise velocities in gas–solid fluid beds are known to be scale dependent[11]; bubbles of the same size rise faster in a column of larger diameter than in columns of a smaller diameter. Analogous scale effects have been found for G-L bubble columns and G-L-S slurry columns and it has been shown that a unified model can be used to estimate the 'dilute' phase hold-up[3,5]. This unified model is based on

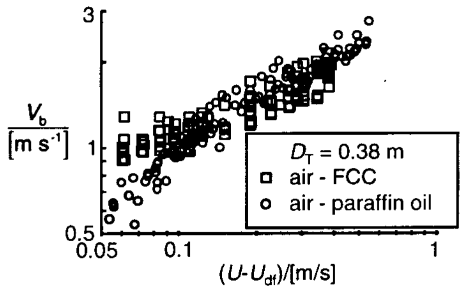

Figure 6. The rise velocity of the "dilute' phase in G-S fluid bed and G-L bubble column; Data obtained in column of 0.38 m diameter with air-FCC and air-paraffin oil.

the bubble growth model of Darton, Davidson and coworkers[17]. The essential idea is that the fast-rising 'dilute' phase (= 'bubbles' for G-S; = 'large bubbles' for G-L and G-L-S systems) is formed by coalescence of smaller bubbles. The coalescence process is limited to an equilibration height h^* above the distributor where the large bubbles reach their equilibrium size. The gas hold-up of the 'dilute' phase for a dispersion height H is

$$\varepsilon_b = \frac{1}{H}\int_0^{h^*} \frac{(U - U_{df})}{V_b}\, dh + \frac{1}{H}\int_{h^*}^{H} \frac{(U - U_{df})}{V_b}\, dh.$$

The rise velocity of the 'large' bubbles, V_b is given by the relation

$$V_b = \phi_0 D_T^n \sqrt{g d_b},$$

taking account of the influence of the column diameter on the rise velocity. The bubble diameter in the growth zone $0 - h^*$ is given by the Darton *et al.*[17] model to be $d_b = \alpha_1\,(U - U_{df})^{2/5}\,(h + h_0)^{4/5}\,g^{-1/5}$. Analytic integration gives the following expression:

$$\varepsilon_b = \frac{1}{\sqrt{\alpha_1}\phi_0 D_T^n g^{2/5}} \frac{\left[(h^* + h_0)^{3/5} - (h_0)^{3/5}\right]}{(3/5)} \frac{(U - U_{df})^{4/5}}{H} + \frac{1}{\sqrt{\alpha_1}\phi_0 D_T^n g^{2/5}} (h^* + h_0)^{-2/5}(H - h^*) \times \frac{(U - U_{df})^{4/5}}{H} \quad \text{for } H \geq h^* \qquad (1)$$

The model parameters, h^*, n and ϕ_0 for air-FCC, air-water and air-paraffin oil are shown in Figure 7. The scale dependence is analogous, but G-L systems display a somewhat weaker dependence on scale. The equilibration height h^* has a much lower value for G-L systems than for G-S systems. The dependence of the 'dilute' phase hold-up on the column diameter is demonstrated in Figure 8, which also shows the success of equation (1) to predict the scale effects in both G-S and G-L systems adequately.

The 'dilute' phase hold-up is found to be practically independent of the liquid phase properties and equation (1)

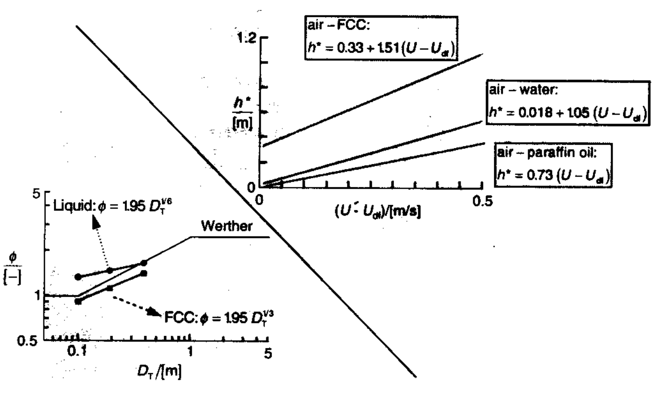

Figure 7. The Werther rise velocity constant and the equilibration height in G-S and G-L systems.

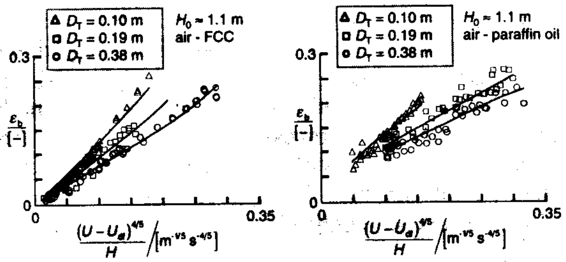

Figure 8. Influence of column diameter on 'dilute' phase holdup.

can be used to predict the 'dilute' phase holdup in viscous liquids and in slurry systems.

DENSE PHASE BACKMIXING

In the heterogeneous flow regime the fast-rising 'dilute' phase tends to churn up the system by creating eddies. The velocity of circulation of the eddies can be expected to be related to the rise velocity of the dilute phase, V_b. The maximum size of the eddy is limited by the size of the vessel, D_T. If we adopt the axial dispersion model for the dense phase backmixing, we can write: $D_{ax} = KV_bD_T$, where K is the constant of proportionality, to be determined experimentally. The rise velocity can be predicted from

$$V_b = \frac{(U - U_{df})}{\varepsilon_b}.$$

Since the rise velocity $V_b \propto D_T^{0.33}$ for G-S fluid beds and $V_b \propto D_T^{0.167}$ for G-L bubble columns (cf. Figure 7), we should expect $D_{ax} \propto D_T^{1.33}$ for G-S fluid beds and $D_{ax} \propto D_T^{1.167}$ for G-L bubble columns. This dependence of the axial dispersion coefficient on the column diameter is in broad agreement with the correlation put forward by Baird and Rice[18]. $D_{ax} = 0.35[g(U - U_{df})]^{1/3}\, D_T^{4/3}$. Taking $K = 0.1$, we find that the predictions of the axial dispersion coefficient are almost indistinguishable from the Baird and Rice correlation.

MASS TRANSFER FOR DILUTE TO DENSE PHASE

In the heterogeneous flow regime the interphase mass transfer between the dilute and dense phases is important in determining the reactor conversion. For gas-solid fluid beds with fine particles, the mass transfer from bubbles is by two mechanisms, convection (through-flow) and diffusion[19]. Though mass transfer in bubble columns have been studied extensively[6,20,21] a clear picture for mass transfer from large bubbles is yet to emerge. In the light of the analogy arguments presented in this paper we may wonder if there is a mechanism for mass transfer from large bubbles equivalent to through-flow in fluid beds; see Figure 9. This through-flow mechanism could involve exchange of gas between large and small bubbles, a much more effective mass transfer mechanism than due to molecular diffusion. Small bubbles could be entrained into the wake of large bubbles and get sheared off at the top, resulting in a convective contribution. This aspect needs to be checked experimentally.

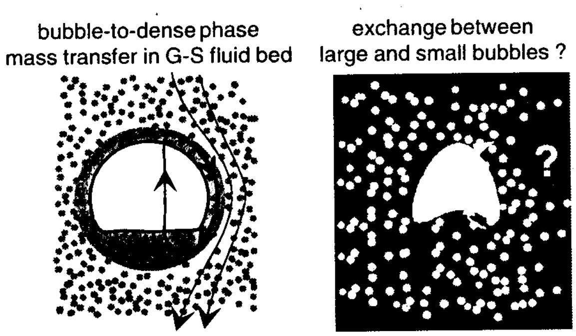

Figure 9. Is there exchange (cross-flow) from large bubbles and small bubbles in G-L bubble columns?

CLOSURE

In this paper we have stressed various hydrodynamic analogies in the operation of G-S fluid beds, G-L and G-L-S bubble columns and suggested a unified model for prediction of the reactor performance. The 'two phase theory' can be extended to handle multiphase systems. The 'dilute' phase can be modelled in a unified manner. The rise velocity of the 'dilute' phase in G-S, G-L and G-L-S systems are of comparable magnitude and exhibit similar scale dependence. Further, experimental data show the 'dilute' phase holdup for G-L and G-L-S systems to be practically independent of the liquid or slurry properties; this is a useful and convenient result because the fast-rising 'dilute' phase dictates gas phase conversion.

Further studies are required to extend the analogous treatment to include mass transfer from the 'dilute' phase.

NOMENCLATURE

d_b	bubble diameter of dilute phase, m
d_b^*	equilibrium bubble size of dilute phase, m
D_{ax}	axial dispersion coefficient, m^2 s^{-1}
D_T	column diameter, m
g	acceleration due to gravity, 9.81 m s^{-2}
h	height above the gas distributor, m
h^*	height above the gas distributor where the bubbles reach equilibrium, m
h_0	parameter determining the initial bubble size, $h_0 = 0.03$ m for porous plate distributors
H, H_0, H_1	height of expanded bed, ungassed bed and after escape of dilute phase, m
K	constant of proportionality in the axial dispersion coefficient correlation
U	superficial gas velocity, m s^{-1}
$(U - U_{df})$	superficial gas velocity through the dilute phase, m s^{-1}
U_{df}	superficial velocity of gas through the dense phase, m s^{-1}
U_{mb}	superficial velocity at which the first 'bubbles' are formed, m s^{-1}
U_{mf}	minimum fluidization velocity, m s^{-1}
U_{trans}	superficial gas velocity at regime transition, m s^{-1}
V_b	rise velocity of the dilute phase, m s^{-1}

Greek letters

α_1	constant, constant in Darton bubble growth model
ε	total gas voidage of G-S or G-L system
ε_b	gas hold-up of 'dilute' phase
ε_{df}	hold-up of gas in 'dense' phase
ε_{mf}	voidage of G-S fluidized bed at minimum fluidization conditions
ε_{trans}	gas hold-up at the regime transition point
ρ_b	bulk density, kg m^{-3}
ρ_p	particle density, kg m^{-3}
ρ_G	density of gaseous phase, kg m^{-3}
ϕ	Werther rise velocity constant, $\phi = \phi_0\, D_T^n$
ϕ_0	constant in rise velocity relationship

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