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AN INVESTIGATION OF THE EFFECT OF BUBBLE DIAMETER ON THE PERFORMANCE OF GAS-SOLID FLUIDIZED BED REACTOR AND TWO-PHASE MODELING OF BUBBLING FLUIDIZED BED REACTOR IN MELAMINE PRODUCTION

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Abstract

Bubble diameter is an important parameter in the modeling of gas-solid fluidized bed reactor and plays an important role in the performance of the fluidized bed reactor. Determination of the actual bubble diameter in the fluidized bed is very important. Bubble diameter starts at the initial bubble diameter (D_{b0}) and grows by coalescence and splitting phenomena, until it reaches the stable bubble diameter at the bed.
Variation of bubble diameter with bed height has been expressed as linear, exponential, and fractional relationships. The linear relationship of Yasui *et al.* [34] predicts the highest value, while the relationship of Park et al. [21] predicts the lowest value and the relationship of Mori and Wen [19] yields an intermediate value. Computation of mean value of bubble diameter is especially important, because it is usually necessary in relationships for determination of bubble velocity. For investigation of the performance of fluidized bed reactor, the two-phase model of Werther [31,32] was used. The results of modeling, including variations of raw material conversion fraction with gas velocity were drawn for various equations of bubble diameter and were compared to experimental conversion fractions.
The results show that the diameter relationships of Mori and Wen [19] and Rowe [23] provide a better fit of data than other equations. For this reason, the equations of Mori and Wen [19] and Rowe [23] are the best relationships for estimation of bubble diameter.
Other relationships, like Darton et al. [5] provide a suitable estimation of bubble diameter in some situations. On the other hand, some equations, like Geldart [8] and Werther [14] do not yield appropriate predictions for bubble diameter.

Keywords: Bubble diameter; Fluidized bed; Modeling; Two-phase; Bubble coalescence; Bubble splitting

1. Introduction

The velocity of mass transport in the bed is a function of bubble size. In general, velocity of transfer between different phases (bubble, cloud, wake, and emulsion) has an important effect on the performance of fluidized bed reactor. For this reason, accurate determination of bubble diameter and its variations with operational conditions and reactor geometry (*e.g.*, diameter and height) is of utmost importance.

In modeling of fluidized bed with constant bubble diameter, the bubble diameter is considered as a parameter of the model. The effective size of bubbles in the bed is determined from experimental data. In this way, it is attempted to fit the experimental conversion fraction with simulated conversion fraction.

Generally, the constant bubble diameter model suffers serious drawbacks for reactor design, scale-up, and optimization, because it uses the average bubble diameter and does not take into account the correlation of bubble diameter with bed height and operational circumstances.

1.1. Bubble Diameter Equations in Gas-Solid Fluidized Bed Reactor

So far, various equations have been proposed for describing variations of bubble diameter through coalescence as a function of apparent gas velocity, design of distributor, and bed height. These equations include linear and exponential functions of bed height (h). These equations have been proposed by Yasui *et al.* [34], Kato and Wen [13], [21] Park *et al.*, Geldart [8], Whitehead *et al.* [33], Rowe *et al.* [23], Darton *et al.* [5], Mori and Wen [19], and Werther [14]. Some of these formulas have been used for fluidized bed; for example, some researchers, including Toor and Calderbank [29], Kato and Wen [13], Mori and Wen [19], and Peters *et al.* [22], have used these equations in their papers. These models have been used primarily for analysis of experimental results and investigation of the effect of operational conditions and model parameters on the performance of fluidized bed. Freyer and Potter [7] conducted studies with the two-phase model of Orcutt *et al.* [20] with plug flow for emulsion (dense) phase using the equations of Kato and Wen [13] for bubble growth.

1.1.1. Frequency of Bubble Coalescence and Splitting in Gas-Solid Fluidized Bed [11]

Two phenomena play a part in the change of bubble diameter: coalescence and splitting. The relative importance of each phenomenon differs for various particles. For large particles of Geldart B, D categories, coalescence causes the change in diameter, while for tiny particles, in addition to coalescence, splitting is also influential in bubble diameter change. Bubble coalescence follows the ideal fluid dynamics, and depends on the diameter of involved bubbles and their elevation (Grace and Clift [4]). On the other hand, frequency of splitting does not depend on the bubble diameter, but is dependent on the size of the particles. Toei *et al.* [27] showed that, according to their study findings, f_S^* is proportional to $dp^{-0.6}$. Tone *et al.* [28] showed that bubble diameter decreases with increasing temperature. This shows that f_S^* is dependent upon the fluid viscosity. Therefore, based on the work by Horio and Nonaka [11], we can express the frequency of bubble splitting with the following formula:

$$f_S^* = f_{S_0} U_{mf}^{-f_{S_1}} \left(f_{S_1} = 1 \approx 1.2 \right) \qquad (1)$$

The effective coalescence frequency is expressed by the following equation:

$$n_{eff} = n_C - n_S \qquad (2)$$

where n_{eff} is effective coalescence frequency, n_C is coalescence frequency, and n_S is bubble splitting frequency. In this equation, n is the frequency in relation with unit height, and it is correlated to bubble production frequency (f_b) according to the following relationship:

$$n_{eff} = n_C - n_S = \frac{-df_b}{dz} \qquad (3)$$

Since bubble production frequency (f_b) is related to bubble diameter (D_b) by

$$f_b = \frac{A\left(U - U_D\right)}{\left(\frac{\pi}{6}\right)\left(D_b^3\right)}, \qquad (4)$$

$\left(n_C - n_S\right)$ is related to bubble diameter (D_b) by

$$\frac{dD_b}{dz} = \frac{\pi D_b^4}{18\left(U - U_D\right)A}\left(n_C - n_S\right) \qquad (5)$$

In these equations, U_D has been considered to include the effect of dense phase expansion in beds with small particles. For beds with large particles, U_D is equal to U_{mf}, while for tiny particles, the $\frac{U_D}{U_{mf}}$ ratio varies between 1 and 4. Anyway, for small particles, the practical gas velocity is greater than the value of U_{mf}, so assuming $U_D = U_{mf}$ will not create a large error.

For particles in B, D categories of Geldart classification, where frequency of bubble splitting (n_S) is insignificant, Equation (5) for computation of coalescence frequency is reduced to the following equation:

$$n_C = \frac{18}{\pi}\left(\frac{(U-U_D)A}{D_b^4}\right)\left(\frac{dD_b}{dz}\right) \qquad (6)$$

The splitting frequency per unit height is expressed by the following simple formula:

n_S = (number of bubble splitting with unit bubble rise) × (bubble production frequency)

$$= \frac{f_S^*}{U_{br}} f_b \qquad (7)$$

By substituting Equation (4) in (7) and using $U_{br} = 0.711\sqrt{gD_b}$, we have

$$n_S = 1.5\frac{(U-U_D)}{D_b^3} D_r^2 \frac{f_S^*}{0.711\sqrt{gD_b}} \qquad (8)$$

Equilibrium bubble diameter (D_{be}) is calculated by assuming the coalescence and splitting frequencies to be equal ($n_C = n_S$).

1.1.2. Bubble Diameter Equations in Gas-Solid Fluidized Bed Reactor for Particles in Geldart B, D Categories

Yasui *et al.* [34] were the first researchers to propose a particle growth formula in which coalescence was taken into account. Their equation is

$$D_b = 0.16\rho_p d_p \left(\frac{U}{U_{mf}} - 1\right)^{0.63} h \qquad (9)$$

Kato and Wen [13] (Kobayashi [15]) entered the initial bubble diameter in the equation for bubble diameter:

$$D_b = 0.14\rho_p d_p \left(\frac{U}{U_f}\right) h + D_{b0} \qquad (10)$$

Geldart [8] expressed the linear correlation of bubble diameter as follows:

$$D_b = 2.05(U-U_{mf})^{0.94} h + D_{b0} \qquad (11)$$

Rowe *et al.* [19]] and Darton *et al.* [5] proposed power functions for bubble diameter:

$$D_b = \frac{0.54(U-U_{mf})^{0.4}\left(h+4\sqrt{A}\right)^{0.8}}{g^{0.2}}$$

(Darton *et al.*) (12)

$$D_b = A_1 + B_1 h + C_1\left(\frac{U}{U_{mf}}\right) + D_1 h\left(\frac{U}{U_{mf}}\right) + E_1\left(\frac{U}{U_{mf}}\right)^2$$

(Rowe *et al.*) (13)

where A_1, B_1, C_1, D_1, and E_1 are constants of the equation.

Rowe *et al.* [23]] also proposed this equation for bubble diameter:

$$D_b = \frac{(U-U_{mf})^{0.5}(h-h_0)^{0.75}}{g^{0.25}} \qquad (14)$$

The value of h_0 depends on the type of plate distributor. For porous distributor, the value of h_0 is zero, while for perforated distributors, its value can be as high as 1 m. The following formula has been proposed for h_0:

$$h_0 = 0.772 + 0.006h \qquad (15)$$

Mori and Web [19] proposed an exponential correlation for bubble diameter:

$$\frac{D_{bm}-D_b}{D_{bm}-D_{b0}}=\exp\left(\frac{-0.3h}{D_r}\right) \qquad (16)$$

where D_r is bed diameter (this equation is notable for considering this parameter), D_{b0} is initial bubble diameter, and D_{bm} is maximum bubble diameter. The precision of this equation is ±50%.

The equation proposed by Mori and Wen [19] is valid for Geldart B, D particles with the following conditions:

$$\begin{gathered} D_r \le 1.3\,\mathrm{m}, \\ 0.005 \le U_{mf} \le 0.2\,\mathrm{m/s}, \\ 60 \le d_p \le 450\,\mu\mathrm{m}, \\ U-U_{mf} \le 0.48\,\mathrm{m} \end{gathered} \qquad (17)$$

Initial bubble diameter (D_{b0}) is an important factor in determination of bubble diameter in the fluidized bed, since the growth of bubble size when it rises from the distributor plate in the bed is dependent on the initial size. Various formulas have been proposed for calculation of initial bubble diameter for various distributor types. One of the most important correlations is the equation presented by Miwa *et al.* [18] for porous, perforated distributor plates. Miwa *et al.* [18] presented a theory about formation of bubbles from a nozzle based on the theory of Davidson and Schulers [19] for a perforated gas distributor:

$$D_{b0}=0.8716\left(\frac{A\left(U-U_{mf}\right)}{ND}\right)^{0.4} \qquad (18)$$

For porous distributor plates, they presented the following equation:

$$D_{b0}=0.376\left(U-U_{mf}\right)^2 \qquad (19)$$

Miwa *et al.* compared the resultant numerical data with experimental findings and found an acceptable fit between them.

Bubbles produced at the top of the distributor plate migrate toward the center of the plate due to the wall effect. This movement of bubbles has been confirmed by data provided by Werther [32]. The radius where maximum bubble volume occurs is a function of bed height. With the rise of bubbles in the fluidized bed, they move toward the center. Bubbles grow in size by coalescence which is caused by increased density of the bubbles toward the center. The terminal state in this process, provided that the bed height is sufficient, is a large bubble that rises along the center of the bed. The diameter of this bubble is designated by D_{bm}. D_{bm} is the maximal bubble diameter that can be achieved through coalescence.

The relationship for maximum bubble diameter is given by:

$$D_{bm}=1.6377\left[A\left(U-U_{mf}\right)\right]^{0.4} \qquad (20)$$

The equation of Mori and Wen is not applicable in the following circumstances:

1) When $D_{bm}>0.3D_r$. In this case, the bed is not a free bubbling bed. Howmand and Davidson [12] showed that in cases where $0.2\le\frac{D_{bm}}{D_r}\le 0.5$, bubbles are in a transitory state between bubbling and slug flow. Therefore, the bubble growth relationship by Mori and Wen is applicable for $D_{bm}<0.3D_r$, until at most $D_b=0.3D_r$; after that, for cases where $D_b>0.3D_r$, the formula for slug flow regime should be used.

2) When the bubble diameter reaches the maximum stable diameter (D_{bs}) before reaching the maximum diameter achievable through coalescence (D_{bm}). Harrison *et al.* [19] proposed this equation for calculation of maximum stable bubble diameter:

$$D_{bs}=\left(\frac{U_t}{0.0071}\right)^2\frac{1}{g} \qquad (21)$$

even though diameters higher than D_{bs} have been observed by some investigators. The discrepancy in findings is partly due to the lack of data on the estimation of limit speed. The above equation is a best guess estimation for the maximum stable diameter (D_{bs}) until better relationships are found. Therefore, the Mori and Wen relationship is applicable for determination of bubble diameter until the diameter reaches the maximum stable diameter ($D_b < D_{bs}$). When bubble diameter reaches D_{bs} , bubbles break down and merge with contiguous bubbles. For this reason, we can say that D_{bs} is always smaller than D_{bm} , and D_{bm} is an artificial diameter which is not observed in reality.

1.1.3. Bubble Diameter Equations in Gas-Solid Fluidized Bed Reactor for Particles in Geldart A Category

If we compute the derivative of the equation by Mori and Wen, we obtain the Equation (22). After substituting in Equation (6), we get the frequency of bubble coalescence for all particle groups.

$$\frac{dD_b}{dz} = 0.3\frac{D_{bm} - D_b}{D_r}$$

(for B, D particles) (22)

$$n_C = 1.35(U - U_D)D_r\frac{D_{bm} - D_b}{D_b^4}$$

(for all groups of particles) (23)

Equation (23) is valid for all groups of particles.

By substituting Equation (23) (for value of n_C) and Equation (8) (for value of n_S) in Equation (5), and integrating for initial values of $D_b = D_{b0}$ and $z = z_0$, we will have the following equation for distribution of bubble diameters along the bed:

$$\left(\frac{\sqrt{D_b} - \sqrt{D_{be}}}{\sqrt{D_{b0}} - \sqrt{D_{be}}}\right)^{1-\gamma_M/\eta}\left(\frac{\sqrt{D_b} + \sqrt{\delta}}{\sqrt{D_{b0}} + \sqrt{\delta}}\right)^{1+\gamma_M/\eta} = \exp\left(\frac{-0.3(z - z_0)}{D_r}\right) \quad (24)$$

where the parameters δ and η and the equilibrium bubble diameter (D_{be}) are given by

$$\frac{D_{be}}{D_r} = \frac{\left[-\gamma_M + \left(\gamma_M^2 + \frac{4D_{bm}}{D_r}\right)^{0.5}\right]^2}{4} \quad (25)$$

$$\gamma_M = \frac{f_s^*}{0.9 \times 0.711}\left(\frac{D_r}{g}\right)^{0.5} \quad (26)$$

$$\frac{\delta}{D_{bm}} = \frac{(\gamma_M + \eta)^2}{4} \quad (27)$$

$$\eta = \left(\gamma_M^2 + \frac{4D_{bm}}{D_r}\right)^{0.5} \quad (28)$$

In conditions of bubble growth where we can ignore splitting and $f_s \to 0$, these parameters approach the following values:

$$\gamma_M \to 0,$$
$$D_{be} \to D_{bm},$$
$$\delta \to D_{bm},$$
$$\eta \to 2\sqrt{\frac{D_{bm}}{D_r}} \quad (29)$$

Equation (24) provides the bubble diameter in gas-solid fluidized bed for particles in A, B, and D groups.

Other equations have been proposed by Darton *et al.* [11] and Rowe [11] that are very similar to the equation by Mori and Wen. Frequency of coalescence according to these relationships have been compared. The results show that the frequency of coalescence is identical in these relationships. The final relationship resulted from Darton *et al.* [11] equation is more complex than Mori and Wen equation (24). For Rowe equation, there is no analytical solution for the integral in Equation (5). For this reason, the results of Equation (24) are preferred for practical applications because of ease of computation.

2. Average Bubble Diameter [2,3]

Effective (average) bubble diameter is generally computed by four ways:

1) Average bubble diameter by integration of the bubble diameter equation:

$$D_{b1} = \int_0^1 D_b(s)ds\ ,\ s = \frac{Z}{H} \qquad (30)$$

2) A bubble diameter that produces a volume expansion identical to bubble diameter change with bubble rise. The value of bubble diameter in this state is given by

$$D_{b2} = \left(\frac{H_{mf}}{H - H_{mf}} \frac{U - U_{mf}}{0.71\sqrt{g}} \right)^2 \qquad (31)$$

where H is given by

$$H = \frac{H_{mf}}{1 - \int_0^1 \varepsilon_b ds} \qquad (32)$$

3) A bubble diameter that creates the same number of mass transfer units as the amount created by bubble change with rising.

$$N_m = \beta H_{mf} \times \left(\frac{4.5 H_{mf} D_{b3}^{0.25} + 5.85 D_e^{0.5} g^{0.25}}{0.71 g^{0.5} D_{b3}^{1.25}} \right) \qquad (33)$$

N_m is the total number of mass transfer units, and is given by

$$N_m = \int_0^1 \left(\frac{K_{be} \varepsilon_b H}{U} \right) ds \qquad (34)$$

4) The value of bubble diameter at height $\frac{H_{mf}}{2}$ or $\frac{H}{2}$:

$$D_{b4} = D_b\left(\frac{H}{2}\right) \text{ or } D_b\left(\frac{H_{mf}}{2}\right) \qquad (35)$$

In this paper, bubble diameter at height $\frac{H_{mf}}{2}$ or $\frac{H}{2}$ is calculated according to bubble diameter equations shown in Table 1, but since the value of height $\frac{H_{mf}}{2}$ is usually known, the bubble diameter at height $\frac{H_{mf}}{2}$ is ordinarily used.

The value of calculated average diameter varies with operational conditions and reactor size. If the effective bubble diameter is known, the other parameters of the model can be calculated. In this situation, the reactor performance will be predicted based on the model in which the parameters do not vary with height. Freyer and Potter [3] proposed the effective bubble diameter D_{b2} as a suitable candidate. Table 1 shows all the equations for computation of bubble diameter in gas-solid fluidized bed.

Werther [3] and Krishna [16] proposed that the effective bubble diameter based on the constant

number of mass transfer units (*e.g.*, D_{b3}) be used. It has been shown that in the absence of resistance for mass transfer between phases, the conversion fraction calculated for steady state using the identical bed expansion is equal to the conversion fraction calculated by the model based on bubble diameter change with height.

Anyway, the exact distribution of bubble diameter variation with bed height is needed only when bubble growth is rapid and the rate of local mass transport decreases intensely with increasing distance from the distributor. Under these circumstances, the model conversion fraction that has been calculated with the assumption of complete mixing in the dense phase, may be higher than the conversion fraction calculated with the assumption of plug flow in the dense phase. This kind of behavior is not seen when using constant bubble diameter model.

3. Modeling of Gas-Solid Fluidized Bed Reactor

We used the two-phase model of Werther [31,32] after some correction for including the effect of various relationships of bubble diameter on the performance of fluidized bed (fluidized bed conversion fraction). The two-phase model presented here is valid for powders in group A and B of Geldart classification (1973)

3.1. Assumptions of Werther Two-phase Model [31,32]

The assumptions for Werther two-phase model are as follows:

1) A distributed bubble phase and the continuous emulsion phase around the bubble constitute the two phases of this model.

2) Since practically $U >> U_{mf}$, the distribution of gas flow in the emulsion phase is low. The velocity range of $\frac{U_{mf}}{\varepsilon_{mf}}$ for gas flow in emulsion phase and $\frac{U_d}{\varepsilon_d}$ for emulsion phase with porosity ε_d is sufficient.

3) Plug flow has been taken into account in both bubble and emulsion phases. (Axial distribution of gas in emulsion phase would be more accurate. For example, De Vries *et al.* considered this in 1972. However, this includes a new parameter, D_{AB}, in the model, the value of which is not known.)

4) Bubbles are void of solid material, and the heterogeneous catalyst reaction or gas-solid reaction takes place on the emulsion phase. Grace and Sun [10], however, showed that there may be a slight amount of tiny particles in the bubbles.

5) Variations of gas volume due to reaction have not been considered. (Sitzmann *et al.* [26] have considered this.)

6) The effects of absorption have been ignored. (RoKita [31] has taken it into account.)

7) This model considers only the bubbling fluidized system.

8) Reaction above the freeboard level has not been considered.

The groups A and B in Geldart classification are defined as follows:

Geldart A particles:

$$\rho_s < 1.4\,\mathrm{g/cm^3}$$
$$d_p < 100\,\mu\mathrm{m}$$

Geldart B particles:

$$1.4 < \rho_s < 4\,\mathrm{g/cm^3}$$
$$40 < d_p < 500\,\mu\mathrm{m} \qquad (36)$$

Table 1. Equations of bubble diameter.

Proponent	Bubble Diameter Equation
Yasui *et al.* (1958)	$D_b = 0.16\rho_P d_p \left(\frac{U}{U_{mf}} - 1\right)^{0.63} h$
Whitehead *et al.* (1967)	$D_b = 9.76\left(\frac{U}{U_{mf}}\right)^{0.33} (0.32h)^{0.54}$
Park *et al.* (1969)	$D_b = 33.3 \cdot 10^3 d_p^{1.5} \left(\frac{U}{U_{mf}} - 1\right)^{0.77} h$
Kato and Wen (1969)	$D_b = 0.14\rho_p d_p \left(\frac{U}{U_{mf}}\right) h + D_{b0}$
Rowe *et al.* (1972)	$D_b = -A_1 + B_1 h + C_1\left(\frac{U}{U_{mf}}\right) + D_1 h\left(\frac{U}{U_{mf}}\right) + E_1\left(\frac{U}{U_{mf}}\right)^2$
Rowe (1976)	$D_b = \frac{\left(U - U_{mf}\right)^{0.5}\left(h + h_0\right)^{0.75}}{g^{0.25}}$ where $h_0 = 0$ for porous plate and $h_0 = 0.772 + 0.006h$ for perforated plate
Geldart (1972)	$D_b = 2.05\left(U - U_{mf}\right)^{0.94} h + D_{b0}$ where $D_{b0} = 0.376\left(U - U_{mf}\right)^2$ for porous plate and $D_{b0} = 0.8716\left(\frac{A\left(U - U_{mf}\right)}{ND}\right)^{0.4}$ for perforated plate. This is valid for group B and D particles.
Mori and Wen (1975)	$D_b = D_{bm} - \left(D_{bm} - D_{b0}\right)\exp\left(\frac{-0.3h}{D_r}\right)$
Werther (1976)	$D_b = 0.00853\left[1 + 27.2\left(U - U_{mf}\right)\right]^{0.333}\left[1 + 6.84h^{1.21}\right]$ for porous plate, where $D_r > 0.2\,\text{m}$, $0.01 \le U_{mf} < 0.08\,\text{m/s}$, $100 \le d_p < 350\,\mu\text{m}$, $0.05 \le U - U_{mf} \le 0.3\,\text{m/s}$
Darton *et al.* (1977)	$D_b = \frac{0.54\left(U - U_{mf}\right)^{0.4}\left(h + 4\sqrt{A}\right)^{0.8}}{g^{0.2}}$

3.2. Modeling Parameters [14,31,32]

The following parameters are included in this model:

1) Bubble diameter (D_b): Bubble diameter equations are based on Table 1 and the results of each correlation for the model will be discussed.

2) Bubble velocity and bubble phase velocity: The absolute velocity of a single isolated bubble was first presented by Davidson and Harrison [6].

$$U_{br} = 0.711\left(gD_b\right)^{0.5} \qquad (37)$$

Davidson and Harrison [6] showed theoretically that the average absolute velocity of aggregated bubbles in the fluidized bed is

$$U_{bo} = U - U_{mf} + U_{br} \qquad (38)$$

According to the findings in several reports, including Werther [31], Werther and Schobler [31], and Hilligardt and Werther [31], the following formula for bubble phase velocity (U_b) was presented:

$$U_b = 0.8\left(U - U_{mf}\right) \qquad (39)$$

3) Bubble phase volume fraction (ε_b): We have

$$\varepsilon_b = \frac{U_b}{U_{bo}} \qquad (40)$$

4) Volume specific mass transfer area: The mass transfer area between bubble and emulsion phases is as follows:

$$\alpha_t = \frac{6\varepsilon_b}{D_b} \qquad (41)$$

5) Coefficient of mass transfer between bubble and emulsion phases (K_{gi}): The bubble and emulsion phase mass transfer coefficient is expressed by Sit and Grace [24] equation:

$$K_{gi} = \frac{U_{mf}}{3} + \sqrt{\frac{4D_i\varepsilon_{mf}U_{bo}}{\pi d_b}} \qquad (42)$$

6) The expanded bed height (H): The bed expands from the height with minimal fluidity. It has been assumed that this expansion is due to the presence of the bubbles. In this case, the bed height is calculated from

$$H = \frac{H_{mf}}{1-\varepsilon_b} \qquad (43)$$

4. Mass Equilibrium Relationships of Werther Two-phase Model [31,32]

Noting the above-mentioned assumptions and condition of mass equilibrium in the reactor volume element $A_t dh$, for a component i in the bubble or emulsion phase, and in the presence of heterogeneous reaction, the following equations are obtained:

$$\varepsilon_b \frac{\partial C_{bi}}{\partial t} = -\left(U - U_{mf}\left(1-\varepsilon_b\right)\right)\frac{\partial C_{bi}}{\partial h} - K_{gi}a_t\left(C_{bi} - C_{di}\right) \qquad (44)$$

in the bubble phase

$$\left(1-\varepsilon_b\right)\left(\varepsilon_{mf} + \left(1-\varepsilon_{mf}\right)\varepsilon_i\right)\frac{\partial C_{di}}{\partial t} = -U_{mf}\left(1-\varepsilon_b\right)\frac{\partial C_{di}}{\partial h} + K_{gi}\alpha_t\left(C_{bi} - C_{di}\right) + \left(1-\varepsilon_b\right)\left(1-\varepsilon_{mf}\right)\rho_s\sum_{j=1}^{M}V_{ij}r_j$$

in the dense phase.

In the steady state, $\frac{\partial C_{di}}{\partial t} = 0$.

As a result, we will have the following equations:

$$\frac{\partial C_{bi}}{\partial h} = \frac{-K_{gi}\alpha_t}{\left(U - U_{mf}\left(1-\varepsilon_b\right)\right)}\left(C_{bi} - C_{di}\right) \qquad (45)$$

$$\frac{\partial C_{di}}{\partial h} = \frac{K_{gi}\alpha_t}{U_{mf}\left(1-\varepsilon_b\right)}\left(C_{bi} - C_{di}\right) + \frac{\left(1-\varepsilon_b\right)\left(1-\varepsilon_{mf}\right)}{U_{mf}\left(1-\varepsilon_b\right)}\sum_{j=1}^{M}V_{ij}r_j \qquad (46)$$

For the reaction rate equation of the i th and j th particle, we have

$$r_j = K_{mj} C_{di}^{nr} \qquad (47)$$

The stoichiometric coefficient of the raw material is 1 ($V_{ij} = -1$), and based on the relationship between K_i and K_{mi}, we have:

$$K_i = \rho_s \left(1 - \varepsilon_{mf}\right) K_{mi} \qquad (48)$$

Therefore, Equation (46) is converted into this equation:

$$\frac{\partial C_{di}}{\partial h} = \frac{K_{gi}\alpha_t}{U_{mf}\left(1-\varepsilon_b\right)}\left(C_{bi} - C_{di}\right) - \frac{K_i}{U_{mf}} C_{di}^{nr} \qquad (49)$$

Equations (45) and (49) are differential equations for variations of concentration in the bubble and emulsion (dense) phases, and they can be solved in the following borderline conditions using the Runge Kutta (fourth order) method:

$$h = 0\ ,\ C_{bi} = C_{di} = C_o \qquad (50)$$

The concentration in bubble and emulsion (dense) phase can be computed for each step (h_1) in the following way:

$$C_b\left(h_1\right) = C_b\left(0\right) + \frac{1}{6}\left(K_1 + 2K_2 + 2K_3 + K_4\right) \qquad (51)$$

$$C_b\left(h_1\right) = C_d\left(0\right) + \frac{1}{6}\left(L_1 + 2L_2 + 2L_3 + L_4\right) \qquad (52)$$

The concentration in all stages is calculated in a similar manner.
The average concentration in any height of the reactor is given by

$$C_{\text{average}} = \frac{C_b U_b + C_d U_{mf}\left(1-\varepsilon_b\right)}{U} \qquad (53)$$

The amount of conversion fraction at any height of the reactor can be computed by

$$x = \frac{1 - C_{\text{average}}}{C_o} \qquad (54)$$

5. Discussion and Comparison of Model Results with Experimental Results

In this section, the results of two-phase model for different equations of bubble diameter have been compared to the published experimental findings on the conversion fraction of first-order isothermal catalyst reaction in gas-solid fluidized bed reactor.

5.1. Ethylene Hydrogenation Reaction

Lewis et al. [17] studied the ethylene hydrogenation reaction. The amount of excess ethylene and reaction is first-order in relation to hydrogen. The catalyst is galvanized cracking nickel. The bed has a diameter of 0.25 m and is kept in temperature of 386 K.

Fluidized gas is distributed from a porous distributor. Some of the data for the system are as follows:

$$D_e = 10^{-4}\ \text{m/s}^2\ ,\ U_{mf} = 0.0073\,\text{m/s}\ ,\ \varepsilon_{mf} = 0.5\ ,\ K = 8.7\,\text{s}^{-1}\ ,\ H_{mf} = 0.25\,\text{m}$$

In Fig. 1, model data for various bubble diameter correlations have been compared to experimental findings. The best fit with experimental results is seen for Mori and Wen, Rowe, Werther, and Darton *et al.* equations, in that order.

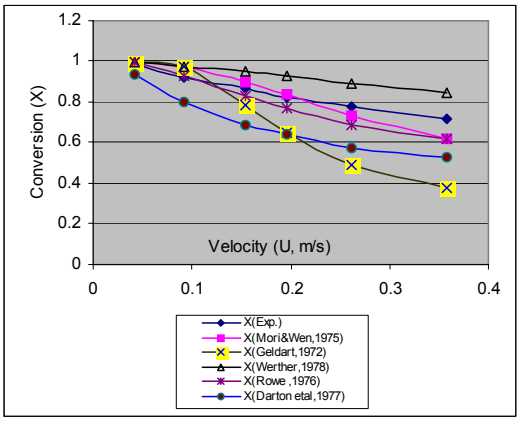

Fig. 1. Comparison of the effect of bubble diameter correlation on fluidized bed conversion. Data of ethylene hydrogenation (Lewis *et al.*, 1959).

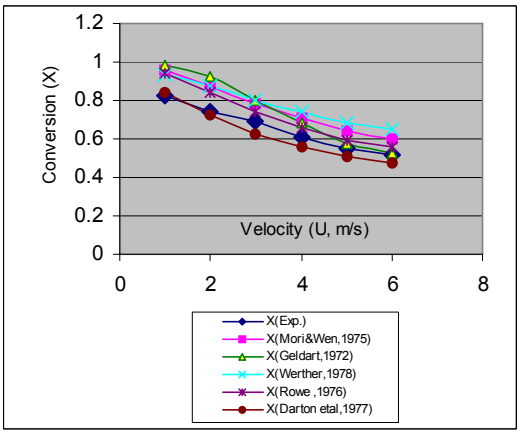

Fig. 2. Comparison of the effect of bubble diameter correlation on fluidized bed conversion. Data of ozone decomposition (Kobayeshi *et al.*, 1967).

5.2. Ozone Decomposition Reaction

a) Kobayashi *et al.* [15] studied the ozone decomposition reaction in a bed with a diameter of 0.83 m. The reaction was performed in the presence of silica gel catalyst in an isothermal manner. Reaction conditions are as follows:

$$U_{mf} = 0.021\,\text{m/s},\ K = 0.7\,\text{s}^{-1},\ H_{mf} = 0.34\,\text{m},\ D_e = 1.8\times10^{-5}\,\text{m/s}^2,\ \varepsilon_{mf} = 0.5$$

The distributor plate is of the porous type.

In Fig. 2, model data for various bubble diameter correlations have been compared to experimental findings. The best fit with experimental results is seen for Mori and Wen, Rowe, Darton *et al.*, Geldart, and Werther equations, in that order.

b) Freyer and Potter [7] conducted the ozone decomposition reaction in a bed with a diameter of 2.92 m (larger than Bench scale). They used sand promulgated with ferrous oxide as catalyst. The data for the reaction was:

$$U_{mf} = 0.017\,\text{m/s},\ \varepsilon_{mf} = 0.48,\ D_e = 1.8\times10^{-5}\,\text{m/s}^2,\ H_{mf} = 0.66\,\text{m},\ K = 3.6\,\text{s}^{-1}$$

The distributor plate is of the perforated type.

In Fig. 3, model data for various bubble diameter correlations have been compared to experimental findings. The best fit with experimental results is seen for Geldart, Rowe, Mori and Wen, Darton *et al.* equations, in that order.

c) Bauer *et al.* [1] studied the ozone decomposition reaction using a bed with a diameter of 0.2 m with a porous distributed plate, a bed with a diameter of 1 m with a perforated distributed plate, and a bed with a diameter of 1 m with a porous-perforated distributed plate. They used sand promulgated with ferric oxide as catalyst (Geldart group B). The data for the reaction was:

$$U = 0.1\,\text{m/s},\ U_{mf} = 0.016\,\text{m/s},\ H_{mf} = 0.5\,\text{m},\ D_e = 1.8\times10^{-5}\,\text{m/s}^2$$

In Fig. 4, model data for various bubble diameter correlations have been compared to experimental findings for the bed with 1-m diameter. The best fit with experimental results is seen for Darton *et al.* and Rowe equations. The other equations, including Geldart, Mori and Wen, and Werther relationships, are in the next positions.

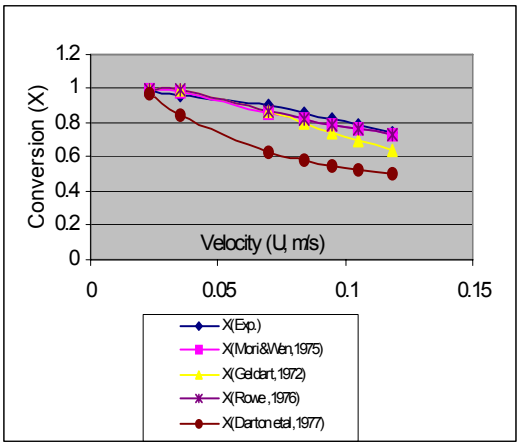

Fig. 3. Comparison of the effect of bubble diameter correlation on fluidized bed conversion. Data of ethylene dehydrogenation (Freyer and Potter, 1976).

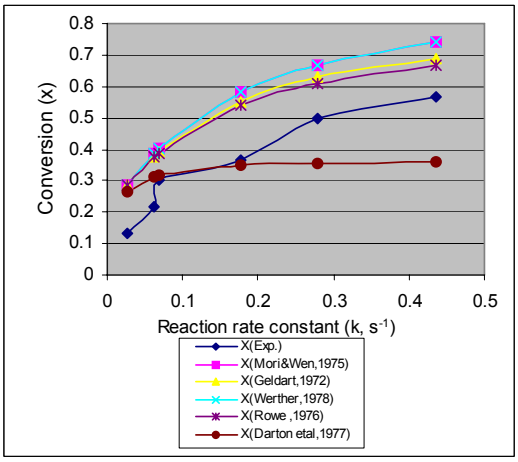

Fig. 4. Comparison of the effect of bubble diameter correlation on fluidized bed conversion. Data of ozone decomposition (Bauer *et al.*,1981).

6. Modeling of Melamine Fluidized Catalyst Bed Reactor Based on Bubble Diameter (Mori and Wen)

Modeling of the melamine production fluidized bed was performed according to the two-phase model of Werther. Hydrodynamic parameters for the fluidized bed with an internal diameter of 4.8 cm and temperature 25°C for silica gel catalyst (weight 400 g) were obtained:

$$H_{mf} = 54\,\text{cm}\,,\; U_{mf} = 2.94\,\text{cm/s}\,,\; W = 400\,\text{g}$$

The physical properties of the catalyst were as follows:

$$\rho_s = 1.96\,\text{g/cm}^3\,,\; \rho_b = 0.43\,\text{g/cm}^3\,,\; d_p = 212\,\mu\text{m}$$

The reaction for production of melamine from urea in the presence of silica gel takes place in temperature 380°C. There is a large volume of ammonia gas in the reaction, so we can assume that ammonia is the fluidizing element of this reaction. The value of minimal fluidity velocity of the particles and minimal bed height vary with increasing temperature.

The equation by Wen and Yu is used for computation of actual minimum fluidizing velocity and yields the actual value of U_{mf}. Based on the diagram of variations of fluidized bed height with velocity, the minimum bed height is calculated. Wen and Yu (1996) presented the following relationship for small particles:

$$U_{mf} = \frac{d_p^2\left(\rho_s - \rho_g\right)}{150\mu} \times \frac{\varepsilon_{mf}^3 \phi_s^2}{1-\varepsilon_{mf}}$$

$$Re_{pmf} < 20 \qquad (55)$$

The relationship for large particles is

$$U_{mf}^2 = \frac{d_p\left(\rho_s - \rho_g\right)}{1.75\rho_g}\varepsilon_{mf}^3$$

$$Re_{pmf} > 1000 \qquad (56)$$

The value of bed voidage at minimum fluidizing state (ε_{mf}) at 380°C is calculated using Broadhurst Becker equation:

$$\varepsilon_{mf} = 0.58\left(\frac{\mu^2}{\rho_g \times g \times \left(\rho_s - \rho_g\right) d_p^3}\right)^{0.029} \left(\frac{\rho_g}{\rho_s}\right)^{0.021}$$

We have

$$\varepsilon_{mf} = 0.425,\ U_{mf} = 2.344\,\text{cm/s},\ H_{mf} = 53.75\,\text{cm}$$

The effective diffusion coefficient of cyanic acid (molecular mass 43) was calculated based on the effective diffusion coefficients of ozone (O_3, molecular mass 46) as follows:

$$D_e = 1\times10^{-5}\ \text{m/s}^2$$

Moreover, since the kinetic data for reaction of melamine production from urea is known, the value of kinetic constant for temperature 380°C is also known.
Reaction rate constant

$$k_{380°C} = 3.4696$$

Order of reaction

$$n = 0.388$$

Therefore, all parameters are ready to be used for modeling.
Since the two-phase model of Werther has been proposed for all orders of the reaction, modeling was performed according to these data. The rate of incoming gas flow was determined according to minimum fluidizing velocity, and values 2 to 10 times U_{mf} were used.

Table 2 shows variations of output conversion fraction of urea according to gas velocity and fluidized bed height based on the two-phase model.

Table 2. Variations of urea conversion fraction with gas velocity and fluidized bed height based on the two-phase model.

U (m/s)	0.2	0.4	0.8	1	1.2
x (two-phase)	0.80	0.61	0.422	0.37	0.33
H_f (m)	0.67	0.77	0.89	0.93	0.97

Fig. 5 shows variations of output conversion fraction of urea according to gas velocity and fluidized bed height based on the two-phase model.

For gas velocity $U = 0.8\,\text{m/s}$, Fig. 6 shows concentration in bubble phase (C_b) and concentration in dense phase (C_d) according to bed height, while Fig. 7 shows variations of urea conversion fraction with bed height.

Fig. 5 shows two-phase model results, including variations of urea conversion fraction and bed height with gas velocity in 380°C.

Fig. 7 shows two-phase model results, including variations of urea conversion fraction with bed height in 380°C.

Fig. 6 shows two-phase model results, including concentration in bubble phase (C_b) and concentration in dense phase (C_d) according to fluidized bed height in 380°C.

Input data for the program are shown in table 3.

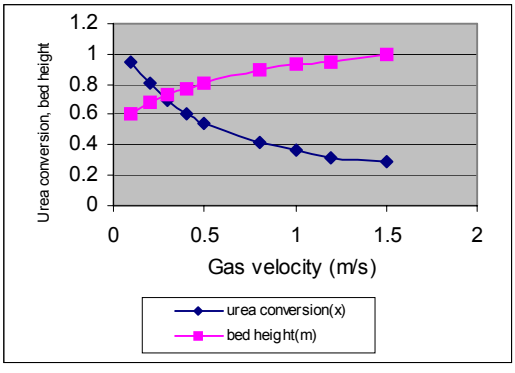

Fig. 5. Two-phase model results: urea conversion and bed height *vs.* gas velocity; U_{mf} = 0.0234 m/s, H_{mf} = .5375 m, D_r = 0.041 m, w = 400 g, ε_{mf} = 0.425, k = 13.7798, n = 0.4, D_e = .00001 m/s^2.

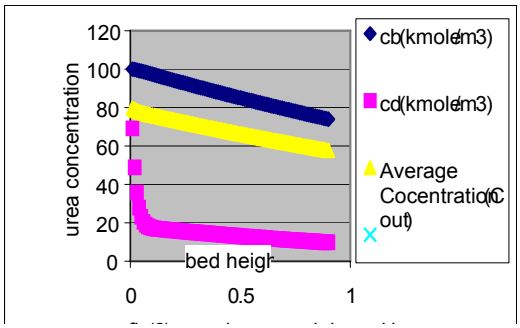

Fig. 6. Two-phase model results: phase concentration profile (bubble, dense, and average) and urea conversion *vs.* bed height; D_r = 4.1 cm, U = 0.8 m/s, U_{mf} = 0.0234 m/s, k = 13.7798, n = 0.4, T = 380°C, D_e = .00001 m/s^2, H_{mf} = .5375 m.

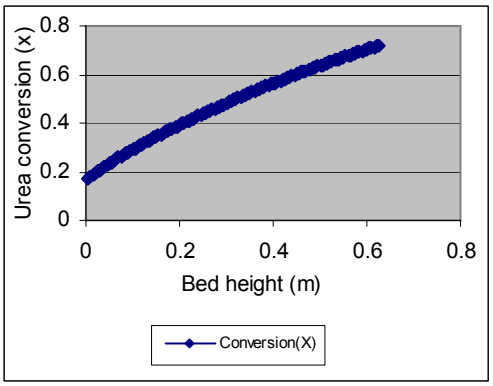

Fig. 7. Two-phase model results: urea conversion *vs.* bed height; D_r = 4.8 cm, U = 0.117 m/s, U_{mf} = 0.02344 m/s, k = 3.4696, n = 0.388, T = 380°C, D_e = 0.00001 m^2/s, H_{mf} = 0.5375 m. Input data for the program are shown below.

Table 3 Performance of Gas-Solid Fluidized Bed Reactor

1	Superficial velocity	U (m/s) := 0.11700
2	Minimum fluidization velocity	Umf (m/s) := 0.02344
3	Particle terminal velocity	Ut (m/s) := 1.00000
4	Length of bed on minimum fluidization condition	Hmf (m) := 0.53750
5	Fluidized bed diameter	Dr (m) := 0.04
6	Distributor type (1: perforated, 2: porous, or 3: Tuyeres)	:= 2.00000
7	The height interval of two phase model	H1 (m) := 0.00100
8	Molecular weight	:= 1.00000
9	Reaction rate constant based on unit volume of dense phase	k (s-1) := 3.46960
10	Initial gas concentration of reactant	C0 (kmole/m3) := 1.00000
11	Bulk density of solid	Sb (kg/m3) := 1.00000
12	Solid density	Ss (kg/m3) := 1.00000
13	Average particle diameter	Dp (m) := 1.00000
14	Gas density	Sg (kg/m3) := 1.00000
15	Average reactor temperature	T (k) := 1.00000
16	Average viscosity of gas	Vg (kg/m.s) := 1.00000
17	Average reactor pressure	P (atm) := 1.00000
18	The order of reaction	nr := 0.38800
19	Gas phase diffusion coefficient	De (m2/s) := 0.00001
20	Bed voidage on minimum fluidization	emf := 0.4250
21	Particle percentage less than 45 micrometer	p45 := 1.00000
22	Number of orifice opening on the distributor	ND := 1.00000
23	Maximum gas velocity on orifice opening	uormax := 1.00000

7. Conclusion

1) Figs. 1 and 3 show that Mori and Wen equation is the best relationship for calculation of bubble diameter, so far. Figs. 2 and 4, however, do not show a satisfactory fit between this relationship and experimental data.

2) The Rowe equation has an apt fitting with experimental results in all cases (Figs. 1 to 4).

3) In pilot scale (Fig. 4), equations of Rowe and Darton *et al.* provide results closer to experimental findings. Other equations are not satisfactory.

4) The reason for the discrepancy between experimental and numerical results is that the average bubble diameter has been calculated as the bubble diameter at height $\frac{H_{mf}}{2}$. This is not a real average value of the bubble diameter and it may not be an accurate approximation of the average

bubble diameter in all circumstances.

5) Rowe and Mori and Wen equations are good formulas for predicting the bubble diameter in gas-solid bubbling fluidized bed reactor. The equation by Darton *et al.* provides good estimates in some cases.

6) Another reason for discrepancy between experimental and model results for conversion fraction, beside the effect of bubble diameter, is the effect of other parameters, like bubble velocity, mass transfer coefficient, and other assumptions of the model. In all cases, appropriate values must be chosen for these parameters.

8-Nomenclature

A	=	Bed area	[m^2]
A_1, B_1, C_1, D_1	=	Coefficients of Rowe *et al.* relationship in Equation (13)	
A_t	=	Volume specific mass transfer area between bubble and emulsion phases	[m^2]
C_{average}	=	Average concentration of gas	[kmol/m^3]
C_o	=	Initial feeding concentration	[kmol/m^3]
C_{bi}	=	Concentration of component i in bubble phase	[kmol/m^3]
C_{di}	=	Concentration of component i in emulsion phase	[kmol/m^3]
D_{bm}	=	Maximum bubble diameter in bed	[m]
D_b	=	Bubble diameter	[m]
D_{be}	=	Equilibrium bubble diameter	[m]
D_{b0}	=	Initial bubble diameter	[m]
D_{b1}	=	Average bubble diameter by integration of Equation (30)	[m]
D_{b2}	=	Average bubble diameter by identical volume expansion, Equation (31)	[m]
D_{b3}	=	Average bubble diameter by identical number of mass transfer units, Equation (33)	[m]
D_{b4}	=	Average bubble diameter as the bubble diameter at height $\frac{H_{mf}}{2}$, Equation (35)	[m]
D_e	=	Gas molecular diffusion coefficient	[m^2/s]
D_r	=	(Reactor) bed diameter	[m]
d_p	=	Particle diameter	[m]
ε_b	=	Bubble voidage	
f_s^*	=	Frequency of bubble splitting	
f_{s0}, f_{s1}	=	Constants of Equation (1)	
g	=	Earth gravitational acceleration	[9.8 m/s^2]
H	=	Bed height	[m]
H_0	=	Initial height where bubble diameter is initial bubble diameter	[m]
H	=	Total bed height	[m]
K	=	Constant for first-order reaction rate	[s^{-1}]
K_{mi}	=	Constant for first-order reaction rate according to catalyst weight	[m^3/kg·s]
K_1, K_2, K_3, K_4	=	Constant values in solution of differential equation with Runge Kutta method	
K_{gi}	=	Coefficient of mass transfer between bubble and emulsion phases for component i	[m/s]
L_1, L_2, L_3, L_4	=	Constant values in solution of differential equation with Runge Kutta method	

N_{eff}	=	Effective coalescence frequency	
n_c	=	Bubble coalescence frequency	
n_s	=	Bubble splitting frequency	
N_m	=	Number of mass transfer units, Equation (33)	
r_j	=	Rate of j th reaction according to catalyst weight	[kmol/kg·s]
t	=	Time	[s]
U	=	Apparent gas velocity	[m/s]
U_b	=	Apparent gas velocity in bubble phase	[m/s]
U_{br}	=	Absolute velocity of single bubble gas	[m/s]
U_{b0}	=	Absolute velocity of bubble aggregate gas	[m/s]
U_{mf}	=	Minimum fluidizing velocity	
U_d	=	Apparent gas velocity in dense phase	[m/s]
U_t	=	Terminal velocity of particles	[m/s]
X	=	Feeding conversion fraction	
Z	=	Height above distributor	[m]
Z_o	=	Initial height where bubble diameter is initial bubble diameter	[m]

Greek Symbols

ε_b	=	Bubble voidage in bed	
ε_{mf}	=	Bed voidage at minimum fluidizing state	
γ_M	=	Parameter in Mori and Wen relationship, Equation (26)	
δ	=	Parameter in Mori and Wen relationship, Equation (27)	
η	=	Parameter in Mori and Wen relationship, Equation (28)	
ρ_p , ρ_s	=	Particle density	[kg/m^3]
ρ_g	=	Gas density	[kg/m^3]
μ	=	Gas viscosity	[kg/m^3]

Indices

m	=	Maximum
S	=	Stable
t	=	Terminal
eff	=	Effective
b	=	Bubble phase
d	=	Dense phase
e	=	Equilibrium
p	=	Particle
r	=	Reactor

References

1. Bauer, W., Werther, J. and Emig, G., 1981, *Ger. Chem. Eng*. 4, 291-2.
2. Bukur, D., B., and Nasif, N., 1985, The effect of bubble size variation on the performance of fluidized bed reactors, *Chem. Eng. Sci.*, 40, 1925-1933.
3. Bukur, D.B., Nasif, N. and Daly, J.G., 1987, On the use of effective bubble diameters in the counter current back-mixing model for fluidized bed reactors, *Chem. Eng. Sci.,* 42, 1510-1513.
4. Clift, R. and J.R. Grace, 1971, Coalescence of Bubbles in fluidized beds, *AICHE Symp. Ser.*, Vol. 67, No. 116, 23.
5. Darton, R.C., R.D. Lanauze, J.F. Davidson and D. Harrison, 1977, Bubble growth due to coalescence in fluidized beds, *Trans. Inst. Chem. Eng.*, 55, 274.
6. Davidson, J. F. and D. Harrison, 1963, *Fluidized particles*, Cambridge, London.
7. Freyer, C. and potter, O.E., 1976, Experimental Investigation of models for fluidized bed catalytic reactors. *AICHE J.*, 22, No. 1, pp. 38-47.

8. Geldart, D., 1971, The size and frequency of bubbles in two and three dimensional gas fluidized beds, *Powder Technology*, 4, 41.
9. Geldart, D., 1973, *Powder technology*, 7, pp. 285-292.
10. Grace, J.R. and sun, G., 1990, powder Technol. G2, 203-205.
11. Horio, M. Nonaka, A., 1987, A Generalized bubble diameter correlation for gas-solid fluidized beds. *AICHE J.*, Vol. 33, No. 11, pp. 1865-1872.
12. Howmand, S. and J. F. Davidson, 1971, Pilot plant and laboratory scale fluidized reactors at high gas velocities; the relevance of slug flow fluidization, ch.5, Academic Press, New York.
13. Kato, and C.Y Wen, 1969, Bubble assemblage, model for fluidized bed catalytic reactors, *Chem. Eng. Sci.*, 24, pp. 1351-1369.
14. Kunii, D. and Levenspiel, D., 1991, Fluidization engineering, 2nd ed., Butterworth-Heinemann, Boston.
15. Kobayashi, H., Arai, F., Izawa, N. and Miya, T. 1967, *Chem. Eng.*, Japan, 5, 28-32.
16. Krishna, R., 1980, *Design and scale-up of gas fluidized bed reactors*. Nato Advanced Study Institute, Vemeiro, Portugal.
17. Lewis, W.K., Gilliland, E.R. and Glass, W., Solid catalyzed reaction in a fluidized bed, *AICHE J.*, 5, 419-429.
18. Miwa, K., S. Mori, S. T. Kato, and T. Muchi, 1970, Behavior of bubble size in large gas-solid fluidized beds, *Chem. Eng.*, Tokyo, 35, 770.
19. Mori, S. and Wen, C.Y., 1975, Estimation of bubble diameter in gaseous fluidized beds, *AICHE J.*, Vol. 21, No. 1, pp. 109-115.
20. Orcutt, J.C., Davidson, J. F. and Pigford R.L., 1962, Reaction time distributions In fluidized catalytic reactors, *Chem. Eng. Prog. Symp. Ser.*, No. 38, 58, 1-15.
21. Park, W.H., Kang W.K, Copes C.E. and Osberg G. L., 1969, The properties of bubble in fluidized beds of conducting particles as measured by an electroresistivity probe, *Chem. Eng. Sci.*, vol. 24, 851.
22. Peters, M.H., Fan, L.S. and Sweeney, T.L., 1982, Reactant dynamics In catalytic fluidized bed reactors with flow reversal of gas in emulsion phase, *Chem. Eng. Sci.*, 37, 553-565.
23. Rowe, P.N., 1976, Prediction of bubble size in a gas fluidized bed, *Chem. Eng. Sci*.
24. Sit, S.P. and Grace. J.R., 1981, *Chem. Eng. Sci.*, 36, 327-335.
25. Sitzmann, W., Werther, J., Bollck, W. and Emig, G., 1985, *Ger. Chem. Eng.*, 8, 301-307.
26. Silzmann, 1986, Dissertation, Echunische Universität Hamburg, Hamburg.
27. Toei, R., Matsono, R., Oichi M. and imamate, K., 1974, Deformations and splittings of a bubble in a two-dimensional fluidized bed: Experimental results, *J. Chem. Eng.*, Japan, 7, 447.
28. Tone, S., Seko, H., Maruyama H. and Otake T., 1974, Catalytic cracking of methylcyclohexane over silica alumina catalyst in gas fluidized bed, *J. Chem. Eng.*, Japan, 7, 44.
29. Toor, F. D. and Calderbank, P. H., 1967, Reaction kinetics in gas-fluidized catalyst beds; Part II: Mathematical models. *Proc. Int. Symp. On Fluidization*, (Edited by Drinkenborgh A.A.H.), pp. 373-392, Netherlands Univ. Press. Amsterdam.
30. De Vries, R.J., van Swaaij, W. P. M., Montovani C. and Heijkoop. A., 1972, *Proc. 5th Europ. Symp. Chem. Reaction Eng.*, Amsterdam, pp. B9 / 59-69.
31. Werther, J., 1992, Scale-up modeling for fluidized bed reactors, *Chem. Eng. Sci.*, Vol. 47, No. 9-11, pp. 2457-2462.
32. Werther, J., 1980, Mathematical modeling of fluidized bed reactors, *Int. Chem. Eng.*, 20. 529-541 .
33. Whitehead, A.B. and young A.D., 1967, Fluidization performance in large scale equipment, Part 1, *Proc. Intern. Symp. On Fluidization*, Eindhoven, Netherlands, pp. 84.
34. Yasui, G. and L.N. Johanson, 1958, Characteristics of gas pockets in fluidized beds, *AICHE*, 4, 445.