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Axial Gas Dispersion in a Fluidized Bed of Polyethylene Particles

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Abstract–Gas mixing behavior was investigated in a residence time distribution experiment in a bubbling fluidized bed of 0.07 m ID and 0.80 m high. Linear low density polyethylene (LLDPE) particles having a mean diameter of 772 μm and a particle size range of 200-1,500 μm were employed as the bed material. The stimulus-response technique with CO_2 as a tracer gas was performed for the RTD study. The effects of gas velocity, aspect ratio (H_0/D) and scale-up on the axial gas dispersion were determined from the unsteady-state dispersion model, and the residence time distributions of gas in the fluidized bed were compared with the ideal reactors. It was found that axial dispersion depends on the gas velocity and aspect ratio of the bed. The dimensionless dispersion coefficient was correlated with Reynolds number and aspect ratio.

Key words: Fluidized Bed, Residence Time Distribution, Gas Backmixing, Axial Dispersion

INTRODUCTION

Gas-solid fluidized beds are among the most important reactor systems in the chemical industry because of their excellent performance in good fluid mixing, high heat transfer rates and low pressure drop in the bed. Fluidized beds are increasingly being used for catalytic polymerization because of a significant reduction in the operating and fixed costs. However, compared with a fluidized bed of FCC particles, the hydrodynamic studies of fluidized bed with polymeric particles are rather limited. An issue yet to be resolved in the modeling of the fluidized bed is whether or not the bed should be treated as a CSTR [Khang and Lee, 1997]. Because bubbling and slugging beds represent severe deviations from the ideal, contacting and the usual procedure of operating a pilot plant and then scaling up is not even completely possible. The main problem lies in the extremely complicated hydrodynamics in the fluidized state, where it is found that large bubbles form in the bed in addition to the existing solid motion. When gas flows through a fluidized bed of solid particles, there is a considerable spread of residence times where different molecules of entering gas spend different lengths of time in passage. This spread of residence times, which is caused by back-mixing, is one of the most important characteristics of a fluidized bed reactor, as it will influence the nature and rate of reaction. Gas back-mixing in a fluidized bed is usually attributed to the downflow of particles. Stephens et al. [1967] pointed out that downflow of gas could occur when the gas velocity of descent of the solids exceeded the interstitial velocity of gas in the dense phase. van Deemter [1961] also mentioned that large aggregates fall down through the bed, carrying with them entrained gas until they are broken up into smaller fragments; small aggregates or single particles are carried upward by the gas stream until they coalesce with other aggregates. Many researchers have studied gas mixing and the patterns of gas flow in fluidized beds, and many models have been proposed to describe the dynamics of a fluidized bed. Most widely used flow models are one-dimensional dispersion models and two-phase models. The former views the bed as a single phase with axially dispersed plug flow of the gas [Edwards and Avidan, 1986], and the latter regards the bed as consisting of a continuous, dense phase containing uniformly dispersed solid particles supported by the fluidizing gas and a discontinuous, bubble phase containing gas bubbles void of high vertical gas velocity [May, 1959; van Deemter, 1961; Kunii and Levenspiel, 1968; Kato and Wen, 1969]. Experimentally, dispersion coefficients are evaluated from two types of tracer studies. First, the unsteady-state experiment injects the tracer in a very short time interval at the entrance of the system and subsequently determines the tracer concentration in the fluid leaving the system [Guo, 1987]. Second, the steady state tracer experiment introduces a steady flow of tracer gas at a horizontal plane in a fluidized bed and measures the upstream diffusion of the tracer [Gilliland and Mason, 1949, 1952; Latham and Potter, 1970]. There is no certainty about the behavior of the gas in this type of reactor owing to the great variety of results and methods used and its variation with the operating conditions. Also, Zacca et al. [1997] demonstrated that residence time distribution effects play a significant role in the establishment of polymer architecture properties such as molecular-weight and average copolymer composition distributions.

In gas fluidized beds, most of the axial mixing is due to the phenomena associated with bubbles, and the motion of the bubbles is affected by the gas velocity and properties of the particles employed. Since the application of fluidized beds for the polymerization reactor has only recently been investigated, research on the flow behavior of polymer particles in a bubbling fluidized bed is limited.

Therefore, in this study a stimulus-response experiment was carried out in order to examine the flow behavior of gas from

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bubbling to just the onset of the turbulent regime with polymeric particles. An unsteady-state dispersion model was used to account for nonideal flow patterns for the whole bed and to determine the axial gas dispersion coefficient, D_a, in the gas phase fluidized bed of polyethylene powder.

EXPERIMENTAL

RTD experiments were carried out in a circular acrylic column with a 0.07 m ID×0.80 m long and disengaging zone of 0.14 m ID×0.20 m long. The experimental test set-up is shown in Fig. 1. Two different bed heights (H_0/D=9.3 and 5.1) were used to examine the effect of aspect ratios on the residence time distribution of gas. The bed was operated at ambient condition and was fluidized with air. The bed material was LLDPE particles with a particle density of 720 kg/m^3 and a mean particle size of 772 μm. The physical properties of the LLDPE particles are given in Table 1. At the beginning of the experiment, the bed was filled to a depth of about 80% by volume of bed with polymer particles. For the RTD test, CO_2 gas was used as the tracer gas. The amount of CO_2 introduced was designed to be 0.1% of the whole bed volume and CO_2 was injected into the bottom of the bed through the copper tube of 0.42 mm ID as an impulse input as shown in Fig. 1. The CO_2 concentration leaving the bed was detected at the exit of the bed by the CO_2 concentration transmitter (Vaisala Co.) which was connected to a computer based data acquisition system. The operating gas velocities were varied from 0.30 m/s to 0.90 m/s corresponding to U/U_{mf}= 2.0 and 6.0. Because the instability of the inlet gas flow may lead to fluctuations in the E(t) curve and the sensor responds sensitively, the tracer concentration at the exit of the bed by voltage was taken by iterating 5-6 times under same conditions before averaging. And then, the measured signals from the probe were smoothed by fast-fourier transform method. In order to examine the scaling up effect on gas mixing in a fluidized bed, the same RTD experiment on a pilot scale with 0.30 m ID×3.50 m height was also carried out.

Table 1. Physical properties of LLDPE particle

	LLDPE particle
Mean diameter	772 μm
Particle density	720 kg/m^3
Voidage at U_{mf}	0.458
U_{mf}	0.154 m/s

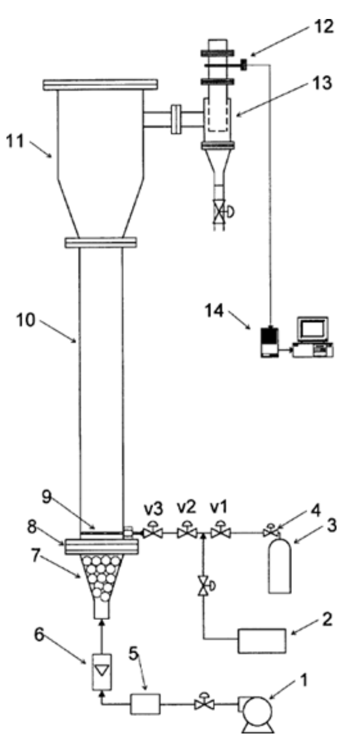

Fig. 1. Schematic diagram of experimental test set-up.

1. Blower
2. Compressor
3. CO_2 reservoir
4. Pressure regulator
5. Filter & Regulator
6. Flowmeter
7. Calming section
8. Distribution plate
9. Tracer injection device
10. Fluidization column
11. Disengaging section
12. Tracer detector
13. Cyclone separator
14. Data analyzer

BASIC THEORY

Residence time distribution (RTD) of gas in a fluidized bed is measured by imposing an idealized instantaneous pulse of tracer on stream entering the vessel at time t=0 and recording the outlet concentration response. At this time tracer concentration is measured by arbitrary unit and injected actual pulse interval must be smaller than about 0.01 times of mean residence time. It is not necessary that the amount of injected tracer be known. Instead, the density function is found by normalizing the outlet response [Nauman, 1981]. To perform this normalization the measured concentration is divided by the area under the concentration-time curve and the normalized response is then called an E curve.

$$E(t)=\frac{C(t)}{\int_0^\infty C(t)dt} \qquad \int_0^\infty E(t)dt=1 \tag{1}$$

The mean residence time and the variance of the response curve are determined as follows.

$$t_m=\int_0^\infty t\,E(t)dt \tag{2}$$

$$\sigma^2=\int_0^\infty (t-t_m)^2\,E(t)dt \tag{3}$$

Li and Weistein [1989] reported that in the low velocity fluidization, including both bubbling and slugging regimes, no significant differences in the radial profiles of concentration were found. Thus, the one-dimensional pseudo-homogeneous axial dispersion model at unsteady-state can be expressed mathematically as

$$D_a\frac{\partial^2 C}{\partial x^2}-\overline{U}\frac{\partial C}{\partial x}=\frac{\partial C}{\partial t} \tag{4}$$

where, $\overline{U}=U/\varepsilon$; interstitial gas velocity

with the following Danckwerts [1953]' closed boundary condition which has no dispersion in the entrance and exit section of the vessel.

$$\begin{aligned} &\text{at } x=0 \quad \bar{U}C_{in}=\bar{U}C(0-) \\ &\text{at } x=0 \quad \bar{U}C_{in}=\bar{U}C(0+)-D_a\frac{dC}{dx}\bigg|_{x\to 0+} \\ &\text{at } x=L \quad \frac{dC}{dx}\bigg|_{x=L}=0,\ C(L-)=C(L+)=C_{out} \end{aligned} \tag{5}$$

The solution obtained in dimensionless form from the above boundary conditions is

$$\Psi(\theta)=\sum_{i=1}^{\infty}\frac{2\delta_i\left(\frac{Pe}{2}\sin\delta_i+\delta_i\cos\delta_i\right)}{\frac{Pe^2}{4}+Pe+\delta_i^2}\exp\left[\frac{Pe}{2}-\frac{\left(\frac{Pe^2}{4}+\delta_i^2\right)\theta}{Pe}\right] \tag{6}$$

where, δ_i=roots of $\cot\delta=\frac{1}{2}\left(\frac{2\delta}{Pe}-\frac{Pe}{2\delta}\right)$, $\Psi(\theta)=\tau\frac{C(t)}{C_{in}}=E(\theta)$

[Levenspiel and Smith, 1957; Wen and Fan, 1975].

The relationship between the Peclet number and the dimensionless variance of RTD can be determined as follows.

$$\sigma_\theta^2=\frac{\sigma^2}{t_m^2}=\frac{2}{Pe}\left[1-\frac{1}{Pe}(1-e^{-Pe})\right] \tag{7}$$

where, $Pe=UH/\varepsilon D_a$

Back-mixing is a flow pattern that is intermediate between the two ideal cases of plug flow and perfect mixing. Axial dispersion model represents plug flow when the dispersion coefficient D_a=0 or Pe=∞. The axial dispersion model lumps the combined effects of fluctuating velocity components, non-flat velocity profiles, and molecular diffusion into the single parameter D_a.

RESULTS AND DISCUSSIONS

The mean residence time of tracer in the fluidized bed as a function of gas velocity for different aspect ratios is shown in Fig. 2. The statistical mean residence time t_m was calculated from the first moment of the RTD such as Eq. (1). From the Fig. 2, it can be seen that the residence time of the gas becomes longer with a tall bed (H_0/D=9.3); this may due to longer contact with downward solid movement in the bed. With increasing gas velocity t_m apparently decreased, and the aspect ratios had a slight effect on the mean residence time of the tracer gas. The effect of bed height on the gas residence time was more obvious from the variance of tracer gas in the fluidized bed.

The variance representing the spread of RTD for different aspect ratio is shown in Fig. 3. As can be seen in Fig. 3, the variance for H_0/D=9.3 was greater than for that for H_0/D=5.1, so more gas-backmixing could be expected to occur in the tall bed. As gas velocity increased, residence time and the spread of RTD were gradually decreased and that was also found in Yates and Constans [1973]. The experimentally determined val-

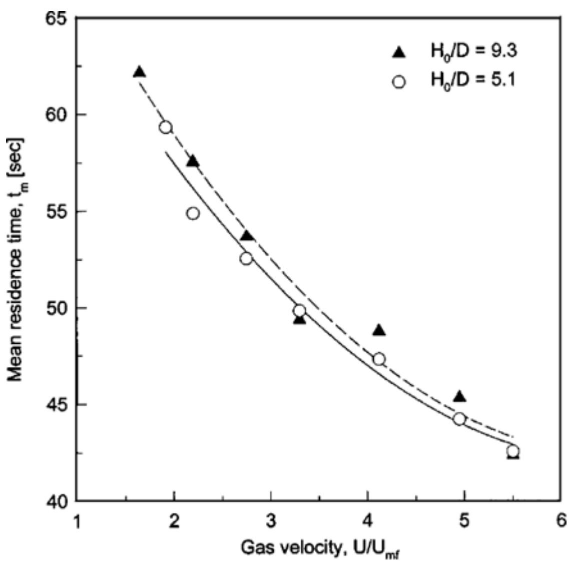

Fig. 2. Mean residence time of trace gas with different aspect ratio.

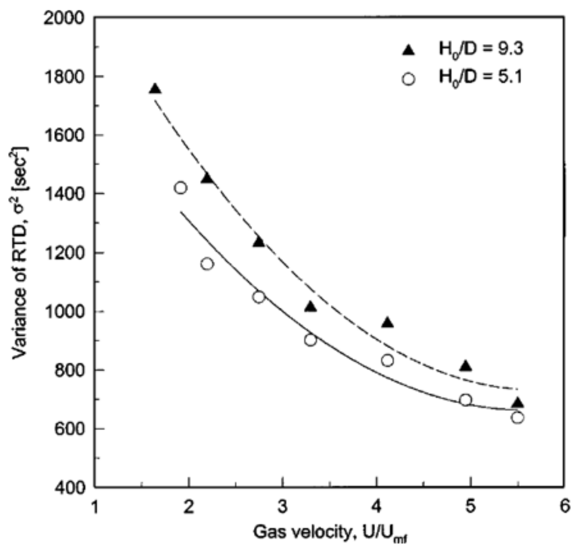

Fig. 3. Variance of tracer gas with different aspect ratio.

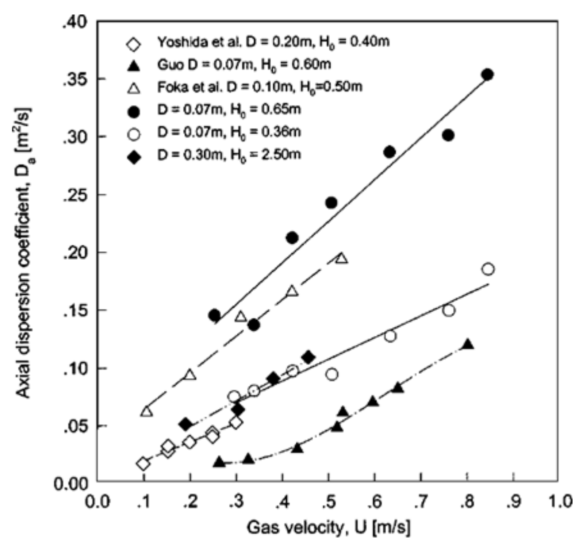

Fig. 4. Experimentally determined axial dispersion coefficient of gas with different aspect ratio.

ues of D_a, as calculated from the experimental Pe number by Eq. (7), were plotted against superficial gas velocity and compared with previous studies [Yoshida et al., 1969; Guo, 1987; Foka et al., 1996] as shown in Fig. 4. The experimental conditions of previous studies are given in Table 2. As shown in Fig. 4, the axial dispersion coefficients, D_a, were in the range of 0.05-0.35 m^2/s for this study and increased with gas velocity. Li and Weinstein [1989] demonstrated that axial dispersion coefficient was almost proportional to the gas velocity until turbulent fluidization began. It can be considered that increasing gas velocity gives rise to more gas back-mixing in the bed by reasons of vigorous solid motion and macro-circulation of solid particles. In agreement with the earlier results of Goedecke et al. [1978] and Guo [1987], it was observed that the experimental unit for the larger aspect ratio had higher axial dispersion coefficients than the smaller one due to longer contact time between solids and gas in the vessel. As shown in Fig. 4, the dependency of the axial dispersion coefficient on the gas velocity was stronger for the larger aspect ratio (H_0/D) than the shorter one. The experimental data of Peclet number obtained in pilot scale shows rather smaller D_a than that of bench scale; then it can be inferred that gas flow behavior in a larger fluidized bed will be closer to plug flow than to complete mixing.

It was also found that the particle size has an effect on the axial dispersion coefficient in the fluidized bed. Guo's [1987] experimental condition, as shown in Table 2, was similar to that of this study except particle size. Guo employed 0.043 mm FCC particles (A type particle) and obtained a smaller axial dispersion coefficient than that of this study with 0.77 mm PE particles (B type particle). This means that larger particles produce more turbulence than smaller ones, thus increasing the gas dispersion in the fluidized bed. This phenomenon was also found from the experimental work of Bang et al. [1999] and Namkung and Kim [1998] in circulating fluidized beds.

In order to propose a simple correlation equation for predicting the axial dispersion coefficient in the fluidized bed of polymeric particles, the dimensionless dispersion group, $D_a\rho_g/\mu$, was introduced and this dispersion group was plotted as a function of particle Reynolds number as shown in Fig. 5. From Fig. 5, it was found that the dispersion group was increased with the particle Reynolds number. Therefore, it can be concluded that D_a was strongly dependent on the particle Reynolds number and the bed dimensions. The experimentally determined dispersion group was correlated with pertinent dimensionless group including par-

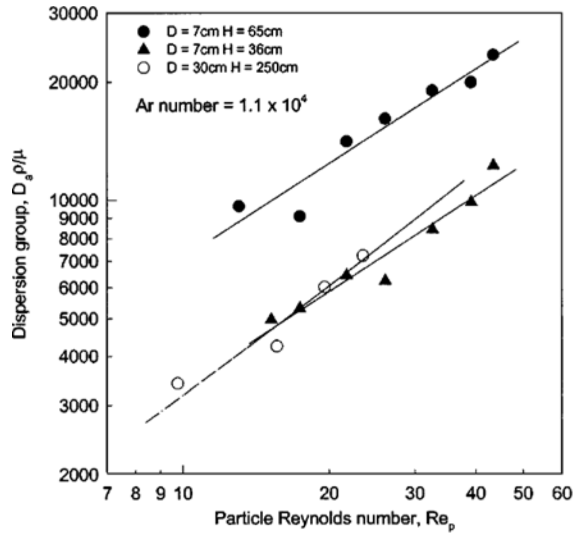

Fig. 5. Effect of particle Reynolds number on axial dispersion coefficient with different aspect ratio.

ticle Reynolds number and aspect ratios. By the non-linear regression method, the following correlation for axial dispersion coefficient in a fluidized bed of polymer particle was proposed with a standard deviation of 5%.

$$D_a\rho_g/\mu = 410.27\,Re_p^{0.81}(H_0/D)^{1.25}(D/d_p)^{-0.40} \tag{8}$$

The proposed correlation showed a good agreement with the experimental data of bench and pilot scale as shown in Fig. 6. Since this proposed correlation was obtained from the polymer particles (Type B) of this study, the validity of the proposed correlation was examined for the other experimental data. Fig. 7 shows the comparison of the proposed correlation with experimental data of this study and previous studies as given in Table 2. As can be seen in Fig. 7, the proposed correlation overestimated the axial dispersion coefficients for type A particles. This discrepancy may come from the difference of particle size. As mentioned before, the larger particles produce more turbulence than the smaller ones, thus increasing the gas dispersion in the fluidized bed [Namkung and Kim, 1998; Bang et al., 1999]. Also, the effect of aspect ratio (H_0/D) on the axial dispersion of gas is shown in Fig. 8. As expected from the proposed correlation, the larger aspect ratio showed larger axial dispersion coefficient than the smaller one. Therefore, it can be said

Table 2. Summary on experimental conditions of previous studies

Authors	Particles	d_p [μm]	ρ_s [kg/m^3]	D [m]	H_0 [m]	Regime	Tracer gas	D_a [m^2/s]
Yoshida et al. [1969]	FCC	60		0.20	0.40	B	He, Freon	0.01-0.23
	Catalyst	150						
Guo [1987]	FCC	43.7		0.07	0.60	B, S, T	H_2	0.015-0.12
Li and Weinstein [1987]	Catalyst	59	1450	0.152		B, S, T, F, Tr	He	0.10-0.78
Foka et al. [1996]	FCC	75	1450	0.10	0.50	B, T	Ar	0.05-0.20
	Sand	130	2650	0.20				
This work	PE	772	720	0.07	0.65	B, S, T	CO_2	0.05-0.35

Note: B: Bubbling; S: Slugging; T: Turbulent; F: Fast; Tr: Transport

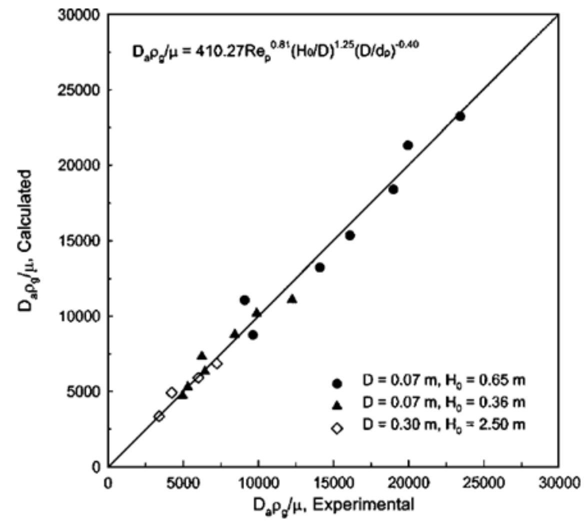

Fig. 6. Comparison of proposed correlation and experimental data of polymer particles for axial dispersion coefficient in a fluidized bed.

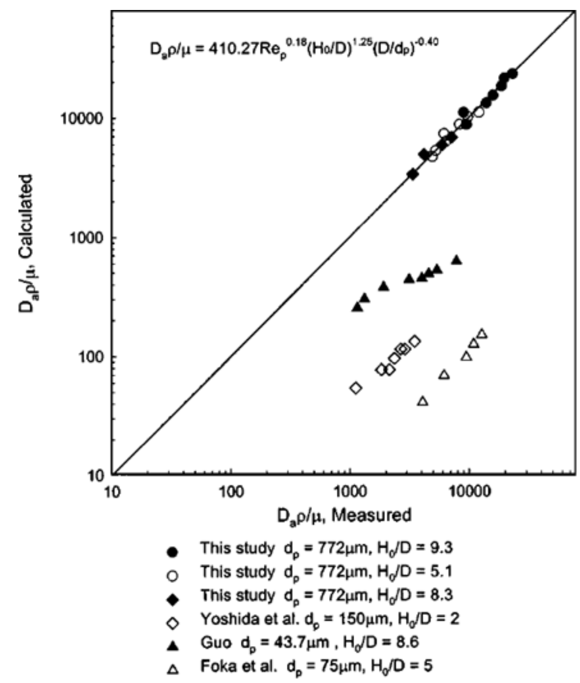

Fig. 7. Comparison of proposed correlation with type A and B particles for axial gas dispersion coefficient in a fluidized bed.

that gas dispersion in the fluidized bed strongly depends on the particle size and aspect ratio as well as gas velocity.

In order to classify the gas flow behavior in a fluidized bed, the internal age distributions for bench and pilot scale experiments were plotted to compare with that of ideal reactors as a function of dimensionless time as shown in Fig. 9. The bench-scale experimental data was closer to the CSTR, while the pilot-scale data was closer to the PFR. The difference of pilot and bench scale data may arise from the characteristics of a bubbling fluidized bed. Unlike in a packed bed, the gas entering into the bed flowed as bubbles. So for the larger diameter bed,

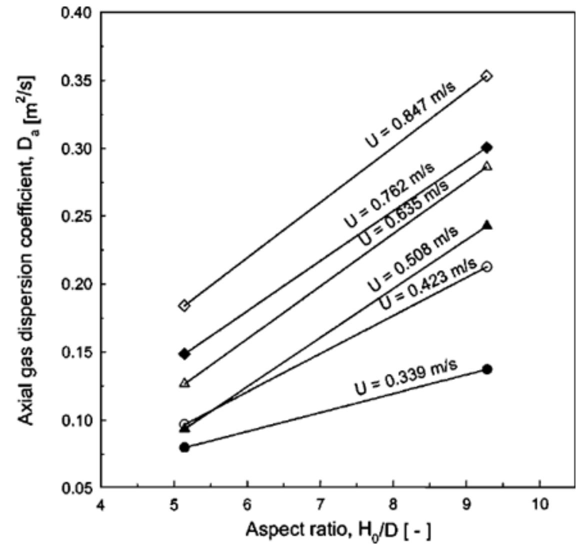

Fig. 8. Effect of aspect ratio on axial dispersion coefficient in a fluidized bed.

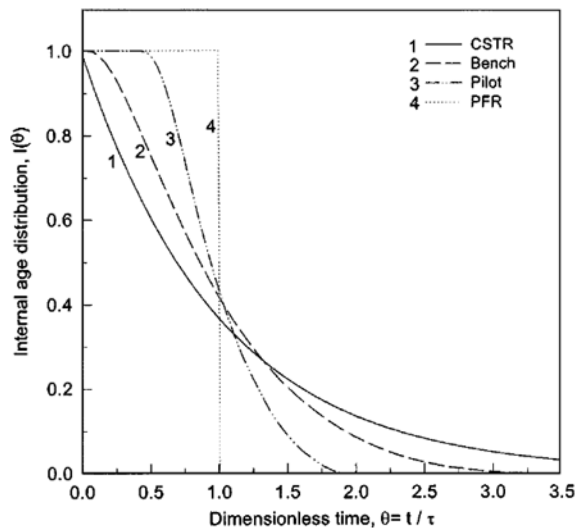

Fig. 9. Comparison of flow behavior of gas in a fluidized bed with ideal reactors.

the bubbles can get bigger and these larger bubbles bypassed without intimate contact with solid particles; thus, gas back mixing is reduced. From Fig. 9, simple plug flow or well-mixed flow assumptions do not model the behavior of gas in the fluidized bed with reasonable accuracy. It can be concluded that the gas behavior in a fluidized bed of polymeric particles lies between CSTR and PFR, and it depends on the particle Reynolds number and bed dimension. The theoretical axial dispersion model was compared with experimental data for different gas velocities of U=0.25 m/s, 0.43 m/s and 0.76 m/s and H_0/D =9.3 in Fig. 10(a), 10(b), and 10(c), respectively. From Fig. 10(a), 10(b), and 10(c), it can be said that at lower gas velocities, the axial dispersion model showed reasonable agreement with experimental results in the fluidized bed of polymer particles and deviated from the dispersion model at the higher gas velocities. It is believed that the deviation from the axial disper-

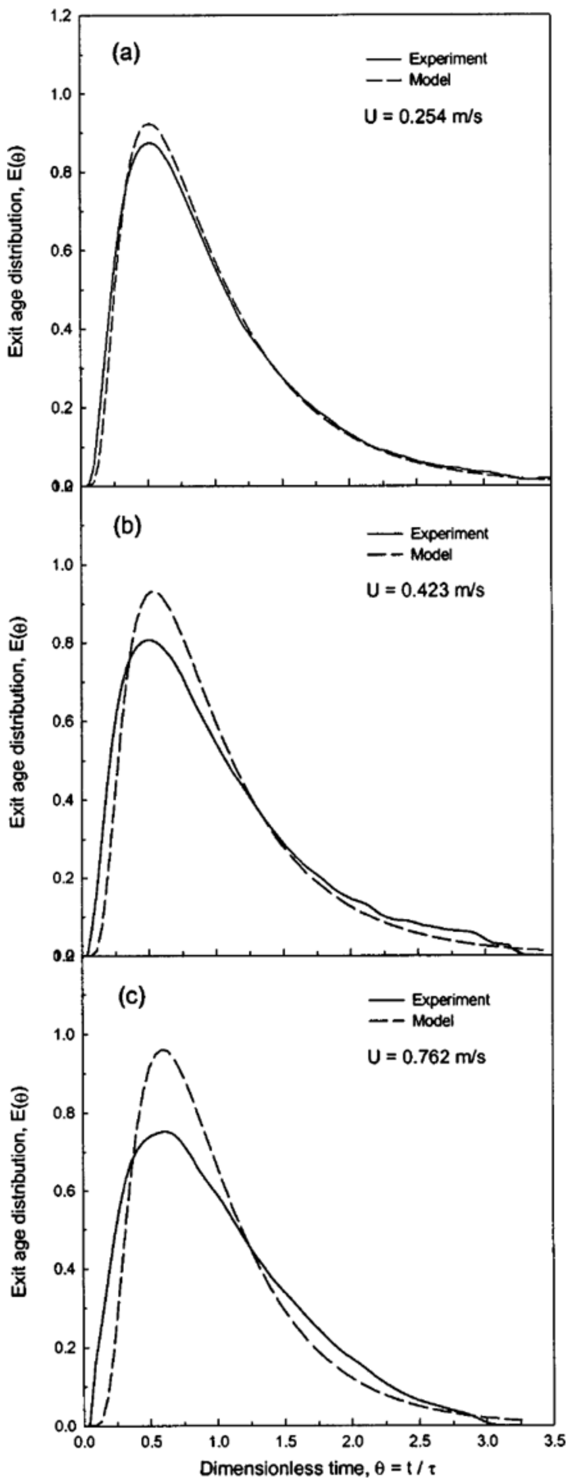

Fig. 10. Comparison of axial dispersion model with experimental results for different gas velocities.

sion model at the higher gas velocities may be due to the flow transition from bubbling bed to slugging bed.

CONCLUSIONS

From the experiment of residence time distribution of gas with a stimulus-response technique in a bench scale and pilot scale fluidized bed of polymeric particles, the following conclusions were obtained.

1. The axial dispersion coefficient increased with the gas velocity and bed height.
2. The flow behavior of gas in a bench scale fluidized bed was closer to CSTR and it changed to the features of PFR as scale-up proceeds.
3. The axial dispersion coefficient was dependent on particle Reynolds number, particle size and aspect ratio.
4. The axial dispersion model of gas in the fluidized bed showed good agreement with experimental data at lower gas velocities and deviated significantly at higher gas velocities.

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NOMENCLATURE

Ar	: Archimedes number, $Ar=d_p^3\rho_g(\rho_s-\rho_g)g/\mu^2$ [-]
C(t)	: concentration of tracer at exit of reactor at time t, arbitrary unit
D	: bed diameter [m]
D_a	: axial dispersion coefficient [m^2/s]
E(t)	: exit-age distribution function [s^{-1}]
E(θ)	: dimensionless exit-age distribution function [-]
H	: expanded bed height [m]
H_0	: static bed height [m]
I(θ)	: internal age distribution function [-]
L	: bed length [m]
$Re_{,p}$	: particle Reynolds number, $Re_{,p}=d_pU\rho_g/\mu$ [-]
Pe	: Peclet number, $Pe=\overline{U}H/D_a=UH/\varepsilon_f D_a$ [-]
Q	: total concentration of tracer entering in the reactor [mol]
U	: superficial gas velocity [m/s]
U_{mf}	: minimum fluidization velocity [m/s]
t_m	: mean residence time [s]

Greek Letters

ε_f	: bed voidage [-]
θ	: dimensionless time [-]
μ	: gas viscosity [kg/m-s]
ρ_g	: gas fluid density [kg/m^3]
σ^2	: variance of residence time distribution [s^2]
σ_θ^2	: dimensionless variance of residence time distribution [-]
Ψ(θ)	: concentration of tracer or exit-age distribution function [-]

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