Korean J. Chem. Eng., **19**(6), 1112-1116 (2002)

Pressure Fluctuation Characteristics of Polyethylene Particles in a Gas-Solid Fluidized Bed with Different Distributors

Dong Hyun Yang, Dong Hyun Lee and Gui Young Han†

Department of Chemical Engineering, SungKyunKwan University, Suwon 440-746, Korea
(*Received 3 May 2002 • accepted 27 September 2002*)

Abstract−The effects of gas velocity (0.10-0.68 m/s), distributor geometry (opening area ratio, number of orifices and hole sizes) and the particle size distribution (wide and narrow) of polyethylene on the pressure fluctuation of the distributor and the local pressure drop in the beds have been determined in a gas-solid fluidized bed of 0.07-m I.D. and 0.7 m in height. The mean amplitude and the normalized standard deviation of the local pressure drop were determined by analyzing pressure fluctuation in the bed. The variation of the mean amplitude of the local pressure drop with the gas velocity is independent on the different distributors. The normalized standard deviation of the local pressure drop increased with increasing the ratio of the excess gas velocity to the minimum fluidization velocity and the lumped parameter of the distributor, h_D. Also, the normalized standard deviation of the local pressure drop of polyethylene polymerized by Metallocene catalyst (narrow) is higher than that of polyethylene polymerized by Ziegler-Natta catalyst (wide).

Key words: Pressure Fluctuation, Polyethylene, Metallocene, Ziegler-Natta, Distributor Types

INTRODUCTION

Polyethylene (PE), which has been manufactured commercially for about seventy years, is now produced in more than forty countries around the world. Among commodity polymers, the annual production of PE is the first. In 1998 the total low-density-polyethylene (LDPE) production was estimated to be about 30 million tons worldwide, and its market demand has been increasing gradually. PE is expected to maintain its position as the first most widely used thermoplastic material in the longer term due to its unique chemical and physical properties. Union Carbide developed a unique and versatile fluidized bed process, called UNIPOL™ process, for producing linear-LDPE (L-LDPE), which is rapidly replacing conventional processes throughout the world [Kunii and Levenspiel, 1991]. The UNIPOL™ process is used in 84 polyethylene reactors and 32 polypropylene reactors worldwide by themselves and licensees [Burdett et al., 2001]. A fluidized bed reactor can be utilized for the gas-phase fluidization of ordinary polymer resin such as L-LDPE since no separation is needed between products and catalyst [Lee et al., 2001]. A large portion of PE has been manufactured in a gas-phase fluidized bed reactor that commonly employs Ziegler-Natta catalyst. Recently, new catalyst has been investigated for good mechanical, chemical and processing properties of PE resins. The homogeneous organometallic catalyst has advantages over Ziegler-Natta catalyst in the polymerization process from much easier control of co-monomers introduction and molecular weight distributions [Cho et al., 2001]. Since physical properties such as particle size distribution, particle shape and particle density are related to properties of the polymerization catalysts, the hydrodynamics of polymer particles in a fluidized bed polymerization reactor would be different with the catalyst type. Only few researches have been carried out regarding the flow behavior of polymer particles in a fluidized bed [Jiang et al., 1994; Cho et al., 2001, 2002; Lee et al., 2001, 2002]. In general, gas-phase olefin polymerization in fluidized beds is operated at a gas velocity of U_g=3.0 to 6.0 U_{mf} and at these gas velocity ranges the flow regime would be bubbling and slugging regimes [Mcauley et al., 1994].

In producing polymer materials in a fluidized bed reactor, bubble size usually increases rapidly to become slugs with increasing gas velocity. Therefore, it is very important to have knowledge of pressure fluctuation in a fluidized bed for gas-phase olefin polymerization.

In the present study, the effects of gas velocity (0.10-0.68 m/s) and distributor geometry (opening area ratio, number of orifices and hole sizes) on the pressure fluctuations including the mean amplitude and the normalized standard deviation of the local pressure drop were determined.

EXPERIMENTAL

Experiments were performed in a fluidized bed (0.07-m I.D.×0.5 m-high) made of a transparent acrylic pipe as shown in Fig. 1 (the details can be found elsewhere [Cho et al., 2002]). The disengaging section (0.14 m-I.D.×0.4 m high) is located between the fluidized bed column and cyclone. Air is supplied from the blower and measured by the calibrated flow meter and introduced to the bottom of the bed through the perforated distributor. Four different types of the gas distributor were employed. In cases of perforated distributors, 400-mesh screen was covered on the surface of a distributor to prevent particle weeping from the bed to the plenum chamber. Details of four different distributors used in this study are shown in Table 1 and Fig. 2. Fig. 2 shows the pressure drop of the four different distributors. As can be seen, the pressure drop of the distributor

†To whom correspondence should be addressed.
E-mail: gyhan@skku.ac.kr
‡This paper is dedicated to Professor Dong Sup Doh on the occasion of his retirement from Korea University.

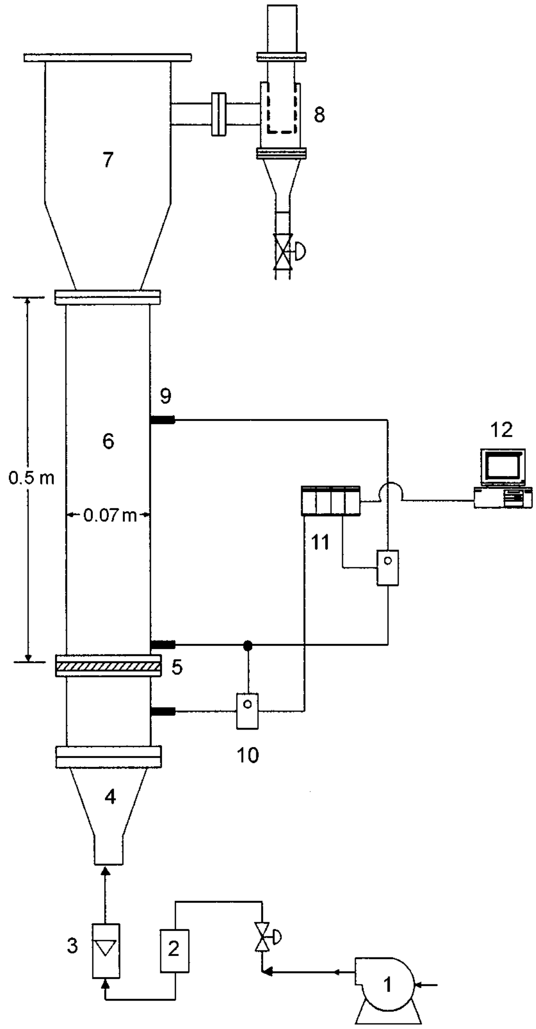

Fig. 1. Schematic diagram of the experimental setup.

1. Blower
2. Filter & Regulator
3. Rotameter
4. Plenum
5. Distributor
6. Main column
7. Disengaging section
8. Cyclone
9. Pressure tap
10. Pressure transducer
11. A/D converter
12. Personal computer

Table 1. Distributor properties for this experiment

Distributor no.	Type	Hole size [mm]	Number [-]	Opening area ratio [%]
1	Perforated	1.0	40	0.8
2	Perforated	1.0	80	1.6
3	Slit nozzle	3.5	6	1.5
4	Perforated	3.0	169	31

increases with increasing gas velocity and decreases with increasing the opening ratio. The physical properties of polyethylene beads used in this work are given in Table 2. The static bed height was about 75% of the bed volume and it was about 0.35 m above the distributor. The pressure drop of the distributor was measured by the pressure taps located between 0.03 m below the distributor and 0.03 m above the distributor. The local pressure drop of the bed was measured through the pressure taps located between 0.03 m and 0.33 m above the distributor. The opening of the pressure taps was covered with a screen of 15 mm aperture to prevent particle leaking from the beds. Transducer signals were processed by a personal computer at a sampling frequency of 100 Hz for 60 s. The operating gas velocity covered 1.0-6.0 U_{mf} and slug flow was ob-

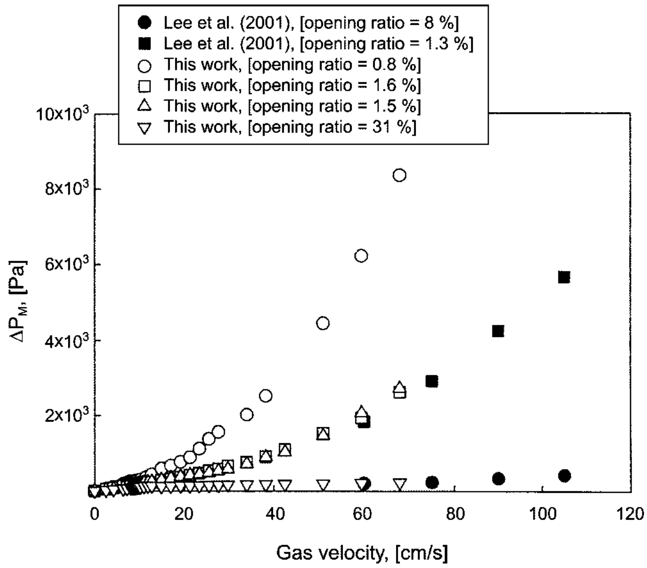

Fig. 2. Effect of gas velocity on the pressure drop across the distributor.

Table 2. Physical properties of Polyethylene beads

Item	Unit	PE of Z-N cat.	PE of Met. cat.
Particle diameter, d_p	μm	593	583
Particle density, ρ_p	kg/m^3	690	830
Particle bulk density, ρ_b	kg/m^3	350	510
Voidage of minimum fluidization, ε_{mf}	-	0.47	0.41
Minimum fluidization velocity, U_{mf}	m/s	0.09	0.10
Particle size distribution	-	wide	narrow

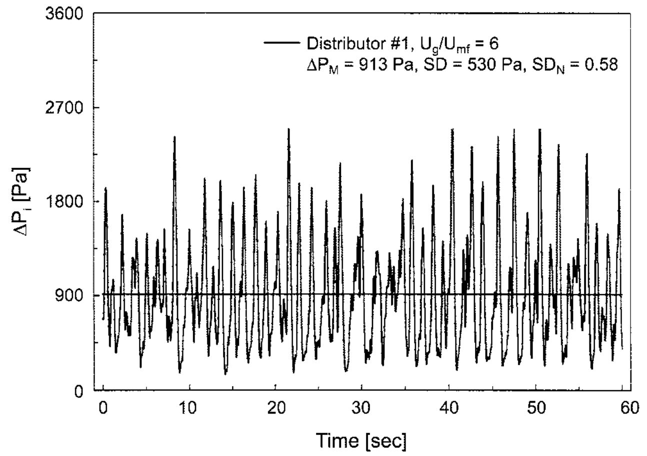

Fig. 3. Typical response signals of the local pressure drop in the beds.

served for that gas velocity range.

Typical pressure signals obtained in the experiments are shown in Fig. 3. The mean amplitude and standard deviation of local pressure drop ($\Delta L=0.3$ m) were calculated by the following equations, respectively:

$$\Delta P_M = \frac{1}{n}\sum_{i=1}^{n}\Delta P_i \tag{1}$$

$$SD = \sqrt{\frac{(\Delta P_i - \Delta P_M)^2}{n-1}} \tag{2}$$

The normalized standard deviation was calculated by

$$SD_N = \frac{SD}{\Delta P_M} \tag{3}$$

As can be seen in Fig. 3, the values of ΔP_M, SD and SD_N are 913, 530 and 0.58, respectively.

RESULTS AND DISCUSSION

The variation of the mean amplitude, ΔP_M, of the local pressure drop ($\Delta L=0.3$ m) with gas velocity in the beds is shown in Fig. 4. As can be seen, the mean amplitude of the local pressure drop in the beds initially increases sharply and then slightly decreases with increasing gas velocity. For low gas velocity in a fixed bed, the mean amplitude of pressure drop is approximately proportional to the gas velocity as reported by the Ergun equation [1952], and usually reaching a maximum, ΔP_{max}. With a further increase in the gas velocity, the mean amplitude of the local pressure drop decreases since the bed voidage increases with increasing the gas velocity beyond U_{mf}. Also, the maximum, ΔP_{max} of the polyethylene polymerized by Metallocene catalyst is higher than that of the polyethylene polymerized by Ziegler-Natta catalyst due to the bulk density of the polyethylene particles as shown in Table 2. However, the variation of the mean amplitude of the local pressure drop with the gas velocity is independent on the different distributors.

The variation of the standard deviation, SD, of the local pressure drop ($\Delta L=0.3$ m) with different types of the distributor in the beds of polyethylene polymerized by Ziegler-Natta catalyst is shown in Fig. 5. In the perforated plate distributor, all the gas in excess of the minimum fluidization velocity forms bubbles of equal size, which are the initial bubbles. The initial bubbles increase with increasing the gas velocity and the orifice hole size. In fluidizing a tall and narrow bed of solids, bubbles formed at the distributor may grow to the bed diameter to form slugs [Kunii and Levenspiel, 1991]. As calculated from the equation of Stewart and Davidson [1967], minimum-slugging velocity, $U_{b,ms}$, of polyethylene polymerized by Ziegler-Natta catalyst is about 0.15 m/s. Therefore, above 0.2 m/s slugging should take place. As can be seen in Fig. 5, the standard deviation of the local pressure drop increased with increasing gas velocity above the minimum slugging velocity.

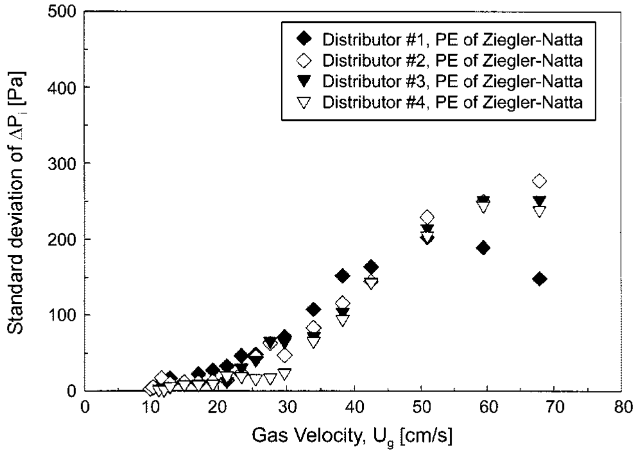

Fig. 5. Standard deviation of the pressure drop in the local bed for different types of distributors.

The effect of the ratio of the excess gas velocity to the minimum fluidization velocity on the normalized standard deviation, SD_N, of the local pressure drop ($\Delta L=0.3$ m) of polyethylene polymerized by Ziegler-Natta catalyst and Metallocene catalyst with different types of the distributor is shown in Fig. 6. As shown, the normalized standard deviation, SD_N, of the local pressure drop increased with increasing the ratio of the excess gas velocity to the minimum

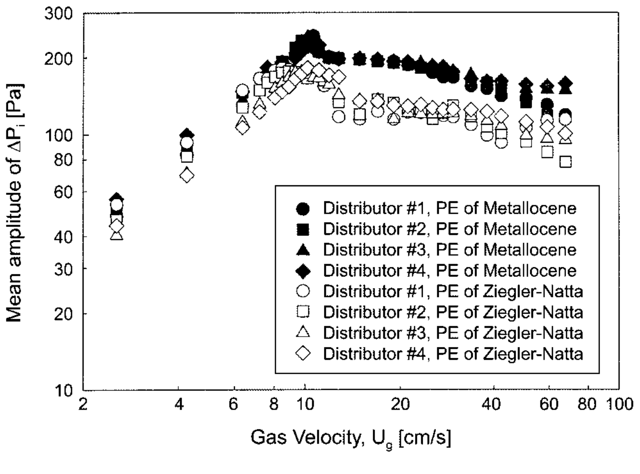

Fig. 4. Mean amplitude of pressure drop in the local bed for different types of distributors.

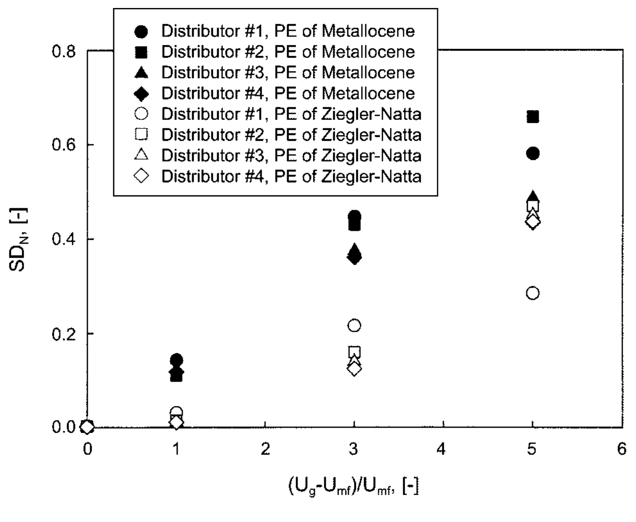

Fig. 6. Effect of the ratio of excess gas velocity to minimum fluidization velocity on the normalized standard deviation.

fluidization velocity due to increasing the initial bubble size. Lee et al. [2001] reported that slug frequency decreases slightly with increasing gas velocity. Also, the normalized standard deviation of the local pressure drop of polyethylene polymerized by Metallocene catalyst is higher than that of polyethylene polymerized by Ziegler-Natta catalyst since the particle size distribution of polyethylene polymerized by Metallocene catalyst is narrow as reported by Cho et al. [2001].

The number of orifice and orifice size in the distributor affect bubble coalescence and slug formations. For air-water systems, Yamashita and Inoue [1975] introduced a distributor parameter as follows:

$$\eta_D = \left(\frac{L_p}{d_{or}}\right)\left(\frac{N_{or}}{D_C^2}\right) = \frac{N_{or}L_p}{d_{or}D_C^2} \tag{4}$$

As reported by Tsuchiya and Nakanishi [1992], this lumped parameter, h_D, could be used for representing the combined effects of the two factors relating to the distributor performance. Lee et al. [2001] also reported that the average slug frequency decreased with increasing η_D with perforated plates.

The effect of η_D on the normalized standard deviation of the local pressure drop with a variation of (U_g/U_{mf}=1 to 6) is shown in Fig. 7. As can be seen, the normalized standard deviation increased slightly with increasing η_D in the beds of polyethylene polymerized by Methallocene catalyst and Ziegler-Natta catalyst due to the decrease of slug frequency as reported by Lee et al. [2001].

CONCLUSIONS

The effects of gas velocity, distributor geometry and the particle size distribution of polyethylene on the pressure fluctuation of the distributor and the local pressure drop in the beds have been determined in a gas-solid fluidized bed. The mean amplitude of the local pressure drop in the bed initially increases sharply and then slightly decreases with increasing gas velocity. The maximum, ΔP_{max} of the polyethylene polymerized by Metallocene catalyst is higher than that of the polyethylene polymerized by Ziegler-Natta catalyst. However, the variation of the mean amplitude of the local pressure drop with the gas velocity is independent on the different distributors. The normalized standard deviation of the local pressure drop increased with increasing $(U_g-U_{mf})/U_{mf}$ and η_D. Also, the normalized standard deviation of the local pressure drop of polyethylene polymerized by Metallocene catalyst (narrow) is higher than that of polyethylene polymerized by Ziegler-Natta catalyst (wide).

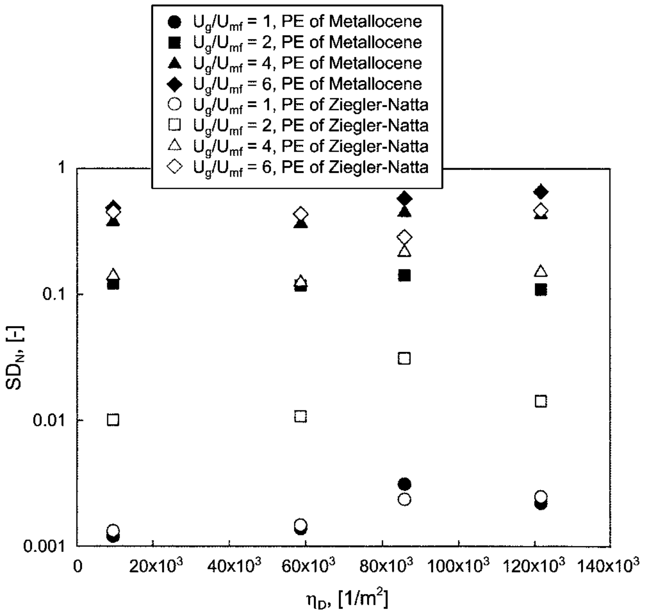

Fig. 7. Effect of η_D on the normalized standard deviation.

NOMENCLATURE

D_c : column diameter [m]
d_{or} : diameter of orifice [m]
d_p : particle diameter [mm]
ΔL : axial distance [m]
L_p : pitched length [m]
n : number of ΔP_i [-]
N_{or} : number of orifice [-]
ΔP_i : instantaneous pressure drop [Pa]
ΔP_M : mean amplitude the local pressure drop [Pa]
ΔP_{max} : maximum pressure drop of the local beds [Pa]
SD : standard deviation of the local pressure drop [Pa]
SD_N : normalized standard deviation of the local pressure drop [Pa]
$U_{b,mb}$: minimum slugging velocity [m/s]
U_g : gas velocity [m/s]
U_{mf} : minimum fluidization velocity [m/s]

Greek Letters

ε_{mf} : voidage of minimum fluidization [-]
η_D : lumped parameter defined by Eq. (4) [1/m^2]
ρ_b : particle bulk density [kg/m^3]
ρ_p : particle density [kg/m^3]

REFERENCES

Burdett, I. D., Eisinger, R. S., Cai, P. and Lee, H. H., "Gas-Phase Fluidization Technology for Production of Polyolefins," Fluidization, X., Kwauk, M., Li, J. and Yang, W. C., eds., United Eng. Found., New York, 39 (2001).

Cho, H., Han, G. and Ahn, G., "Characteristics of Slug Flow in a Fluidized Bed of Polyethylene Particles," *Korean J. Chem. Eng.*, **19**, 183 (2001).

Cho, H., Han, G. and Ahn, G., "The Slug Flow Behavior of Polyethylene Particles Polymerized by Ziegler-Natta and Metallocene Catalysts," *Korean J. Chem. Eng.*, **18**, 561 (2001).

Ergun, S., "Fluid Flow through Packed Columns," *Chem Eng. Prog.*, **48**, 89 (1952).

Jiang, P., Bi, H., Liang, S. C. and Fan, L. S., "Hydrodynamic Behavior of Circulating Fluidized Bed with Polymeric Particles," *AIChE J.*, **40**, 193 (1994).

Kunii, D. and Levenspiel, O., "Fluidization Engineering," 2nd ed., Butterworth-Heinemann (1992).

Lee, S. H., Lee, D. H. and Kim, S. D., "Slug Characteristics of Polymer Particles in a Gas-Solid Fluidized Bed," *Korean J. Chem. Eng.*, **19**, 351 (2002).

Lee, S. H., Lee, D. H. and Kim, S. D., "Slug Characteristics of Polymer Particles in a Fluidized Bed with Different Distributors," *Korean J. Chem. Eng.*, **18**, 387 (2001).

Mcauley, K. B., Talbot, J. P. and Harris, T. J., "A Comparison of Two-Phase and Well-Mixed Models for Fluidized Polyethylene Reactors," *Chem. Eng. Sci.*, **49**, 2035 (1994).

Stewart, P. S. B. and Davidson, J. F., "Slug Flow in Fluidized Beds," *Powder Techn.*, **1**, 61 (1967).

Tsuchiya, K. and Nakanishi, O., "Gas Holdup Behavior in a Tall Bubble Column with Perforated Plate Distributors," *Chem. Eng. Sci.*, **47**, 3347 (1992).

Yamashida, F. and Inoue, H., "Gas Holdup in Bubble Columns," *J. Chem. Eng. Japan*, **8**, 334 (1975).