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Phase Hold-up and Critical Fluidization Velocity in a Three-Phase Inverse Fluidized Bed

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Abstract−We studied the hydrodynamic characteristics of a three-phase inverse fluidized bed made of a transparent acrylic column of 0.115 m inner diameter and 2 m heights. Air, water and polyethylene particles were used as the gas, liquid and solid phase, respectively. We used both hydrophobic low density polyethylene (LDPE) and hydrophilic LDPE as solid phase, and distilled water as liquid phase, and filtered air as gas phase. The LDPE was chemically treated by chlorosulfonic acid to change the surface property from hydrophobic to hydrophilic. We tried to solely investigate the effect of the surface hydrophilicity of polymeric particles on the phase holdup and the critical fluidization velocity of three-phase inverse fluidization. Thus, we measured the static pressure and eventually observed critical fluidization velocity. Critical fluidization velocity became smaller in case of using MDPE hydrophobic particles than LDPE hydrophilic particles. This was thought to be due to the retardation of rising bubbles near hydrophobic particles and, subsequently, the increase of gas hold-up.

Key words: Inverse Fluidized Bed, Hydrophobic, Hydrophilic, Critical Fluidization Velocity

INTRODUCTION

The industrial application of three-phase fluidization has gradually increased in many different chemical processes since the technique has a low-pressure drop and easy controllability of operating variables like temperature and pressure [Kim et al., 1980, 1997; Kang et al., 1995, 1997, 1999; Matsuura et al., 1984]. Since three phases are involved in the process, however, the phenomena in the fluidized bed are so complicated that the performance of the apparatus is hardly predictable. Thus, the investigation of basic hydrodynamic characteristics before application is very important for successful operation. Recently, a new concept of inverse fluidization was developed and actively studied. This system is operated with downward flow of liquid counter to the net upward buoyancy force on the particles. The gas flow is upward, counter to the liquid flow and bed expansion can be supported either by the downward liquid phase or by the upward gas phase, or by both [Fan et al., 1982; Krishnaiah et al., 1993; Buffiere et al., 1999; Uribe-Salas et al., 1994; Briens et al., 1999; Choi et al., 1999; Han et al., 2002; Comte et al., 1997]. The advantage of this technique is low fluid velocity, small energy, and easy fluidization without breakage of solid particles. The essence of this technique is to fluidize floating light particles downward by lowering the density of fluid through the injection of gas from bottom side. To fully understand this phenomenon, the investigation of the hydrodynamic characteristics and the phase holdups during fluidization is a very important first step. So far, many researchers have studied the hydrodynamic characteristics of three-phase inverse fluidization. However, no one was aware of the effect of solid surface properties on the hydrodynamic characteristics of the three-phase inverse fluidization. Thus, the primary objective of this study is to understand how the surface property of solid particles affects the hydrodynamic behavior of inverse fluidization. In addition to this, we compared the effect of surface properties of particles with the effect of density of particles on the hydrodynamic performance of the inverse fluidization. To do this, we carefully inves-

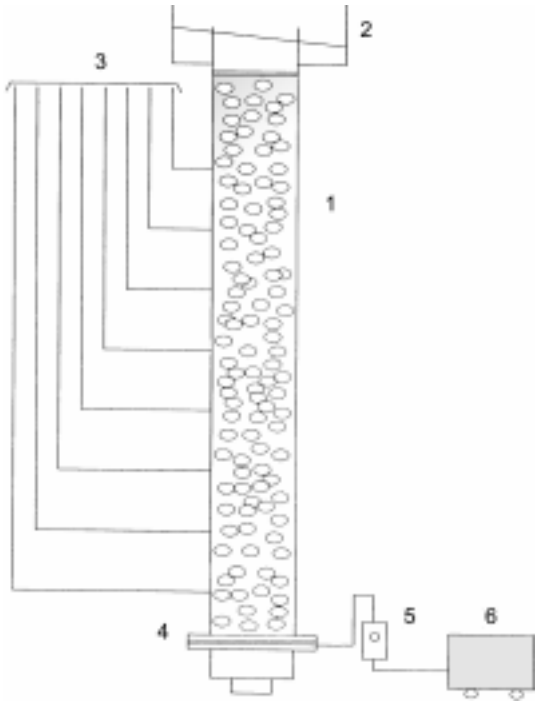

Fig. 1. Schematic diagram of experimental apparatus.
1. Main column
2. Weir
3. Manometer
4. Air-liquid distributor
5. Flow meter
6. Compressor

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‡This paper is dedicated to Professor Dong Sup Doh on the occasion of his retirement from Korea University.

Table 1. Physical properties of LDPE/MDPE particles

	Average diameter (mm)	Thickness (mm)	Average density (kg/m^3)
LDPE hydrophobic particle	4.4	1.6	926
LDPE hydrophilic particle	4.4	1.6	940
MDPE hydrophobic particle	4.4	1.6	940

tigated the change of phase holdups with respect to gas inlet velocity and the change of critical fluidization velocity with respect to particle loads.

EXPERIMENTAL PROCEDURES

Fig. 1 shows the inverse fluidized bed apparatus used in this experiment. The main column of the apparatus is a transparent acrylic column of inner diameter of 0.115 m and height of 2 m. Solid particles used in this experiment were hydrophilic/hydrophobic LDPE and hydrophobic MDPE particles. The hydrophobic LDPE particles and hydrophobic MDPE particles were kindly supplied by HANWHA Co. and SK Co., respectively. Table 1 represents the physical properties of particles. Filtered compressed air, distilled water and polymers were used as the gas, liquid and solid phases, respectively. A perforated plate was used as a gas distributor. Equally spaced pressure taps were mounted on the column wall from the distributor up and connected to water manometers. To prevent solid particles from entering into the pressure measurement lines, a polymeric screen was attached to the tip of each pressure tap.

To prepare hydrophilic LDPE particles, we chemically modified the surface of hydrophobic LDPE particles. First, LDPE particles were mixed with 10 mM-ferric chloride solutions at 30 °C for 3 hours. The pretreated particles were distilled water washed and vacuum dried for 30 min. Then, we etched the particles in chlorosulfonic acid solution [$ClSO_3H : CCl_4$=2 : 1 (w/w)] with stirring for 4 hours at room temperature. After the particles were etched, they were distilled water washed for at least 15 min and vacuum-dried over 1 hr [Yang et al., 1997].

In this experiment, the liquid phase was static and only the gas phase was injected through the bottom distributor. To avoid the possibility of gas packing in case of increasing gas inlet velocity, we performed the inverse fluidization experiment while decreasing gas inlet velocity after fully fluidizing the bed at high inlet velocity.

We measured the average phase holdups in the three-phase fluidized bed from the following equations:

$$-\frac{dP}{dz}=(\varepsilon_G\rho_G+\varepsilon_L\rho_L+\varepsilon_S\rho_S)\frac{g}{g_C} \tag{1}$$

$$\varepsilon_G+\varepsilon_L+\varepsilon_S=1 \tag{2}$$

$$\varepsilon_S=\frac{M_s}{H_{b0}A\rho_S} \tag{3}$$

To analyze our experimental data, we used Comte et al.'s model [Comte et al., 1997]. The main assumption of their model is that the axial solid distribution in the column is mainly controlled by the difference between the density of the particle and that of the gas-liquid mixture, which can be considered as a homogeneous fluid. The density of this mixture can be estimated as follows everywhere in the reactor whatever the conditions are:

$$\bar{\rho}=\frac{\varepsilon_G\rho_G+\varepsilon_L\rho_L}{\varepsilon_G+\varepsilon_L} \tag{4}$$

Since the density of gas is much less than the density of liquid, Eq. (4) can be simplified by

$$\bar{\rho}=\frac{\varepsilon_L\rho_L}{\varepsilon_G+\varepsilon_L} \tag{5}$$

If the density of solid particles is greater than the density of gas-liquid mixture, particles will settle down to the bottom of the reactor. If the density of solid particles is less than the density of the mixture, particles will go upward. When the density of solid particles is equal to the density of the mixture, particles will disperse in the reactor. In this model, however, the liquid circulation due to the rising gas bubbles is not considered. When the gas velocity is equal to the critical fluidization velocity, the density of the mixture equals to the density of solid particles.

That is,

$$\bar{\rho}=\rho_S \tag{6}$$

and Eq. (5) becomes

$$\frac{\rho_S}{\rho_L}=\frac{\varepsilon_L}{\varepsilon_G+\varepsilon_L} \tag{7}$$

If we define R as

$$R\equiv\frac{\rho_L-\rho_S}{\rho_L}=1-\frac{\rho_S}{\rho_L} \tag{8}$$

and combine Eqs. (2), (7) and (8), we have

$$\varepsilon_L=(1-\varepsilon_S)\frac{\rho_S}{\rho_L} \tag{9}$$

and

$$\varepsilon_G=R(1-\varepsilon_S) \tag{10}$$

From the concept of the bubble swarm velocity G defined by Wallis [Wallis et al., 1969],

$$G=\frac{U_{Gc}}{\varepsilon_G} \tag{11}$$

the critical fluidization velocity can be expressed as follows:

$$U_{Gc}=GR(1-\varepsilon_S) \tag{12}$$

RESULTS AND DISCUSSIONS

1. Phase Holdups

Figs. 2-4 represent the change of phase holdups at three phase region with respect to the change of gas inlet velocity. Solid holdups gradually decreased due to the bed expansion with increasing gas inlet velocity. When the solid holdup becomes constant, particles are uniformly distributed in the whole fluidized bed. The gas inlet velocity at this point was defined as the critical fluidization velocity, U_{GC}. To decide the critical point, we carefully observed the uniform distribution of particles in the bed with slightly changing the gas inlet velocity from 0.01 to 0.06 m/sec. At each experi-

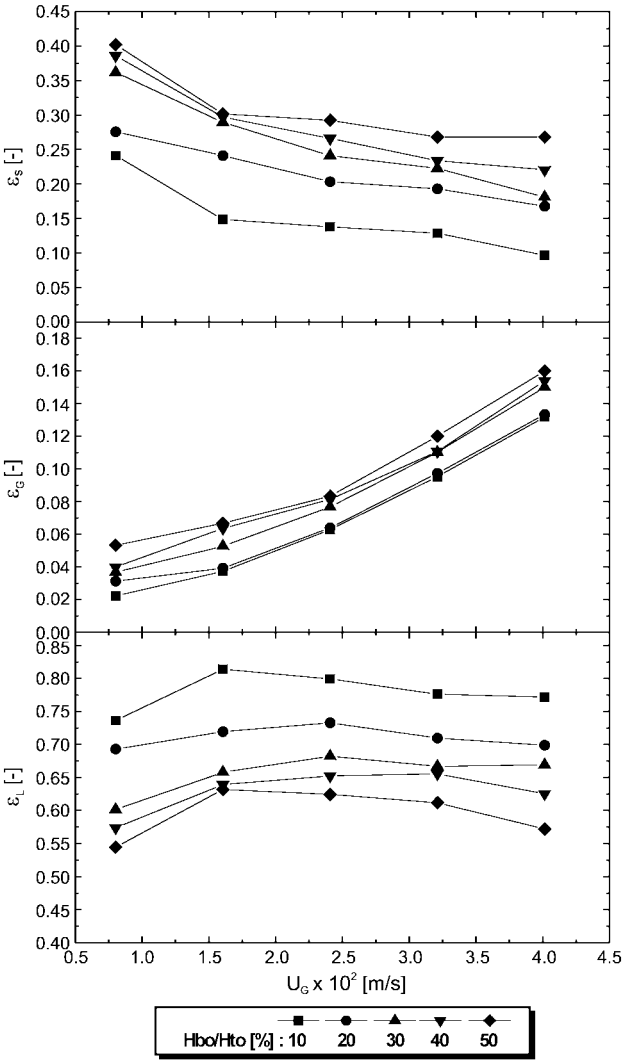

Fig. 2. Effect of gas inlet velocity on the individual phase holdups in three-phase inverse fluidized beds of LDPE hydrophobic particles.

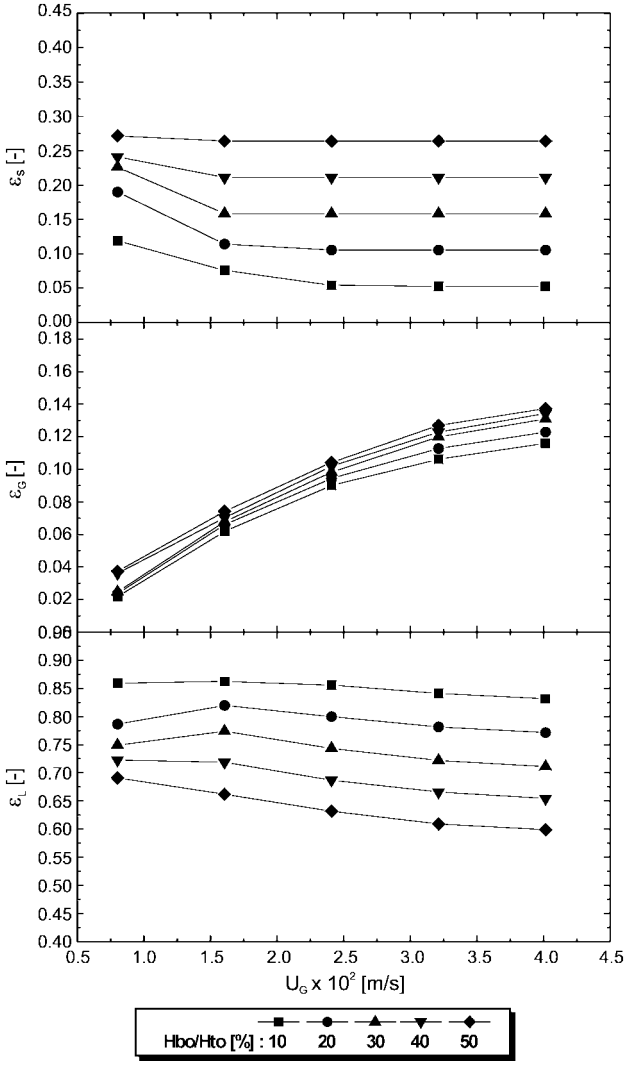

Fig. 3. Effect of gas inlet velocity on the individual phase holdups in three-phase inverse fluidized beds of LDPE hydrophilic particles.

ment, we also changed the particle loads (H_{b0}/H_{t0}) from 10 to 50%.

Fig. 2 represents the change of phase holdups when we use hydrophobic LDPE particles (ρ_S=926 kg/m^3). While solid holdups decreased with increasing gas velocity, gas holdups exponentially increased and liquid holdups showed a local maximum. Fig. 3 shows the change of phase holdups when we use hydrophilic LDPE particles (ρ_S=940 kg/m^3). In this case, solid holdups became easily constant even at small gas velocity and gas holdups monotonically increased with increasing gas velocity. Fig. 4 shows the case of hydrophobic MDPE particles (ρ_S=940 kg/m^3). The behaviors of phase holdups are the same as those of hydrophilic LDPE particles but gas holdups are slightly larger.

Since the hydrophobic LDPE particles have lower density than other particles and the gas velocity is far below U_{GC}, solid holdups are still decreasing even at high gas holdups except the cases of 40 and 50% (Fig. 2). And the gas holdups are concavely increasing with increasing gas inlet velocity unlike the convex shapes of Figs. 3 and 4. This concave pattern results in the local maximum of liquid holdups. Since the three-phase region is not fully expanded under the condition of this low gas velocity, upcoming bubbles are temporarily stopped at the interface between two-phase and three-phase and coalesced into slugging form before penetrating the three-phase region. This phenomenon was observed especially at small particle loads and low gas velocity. As the gas velocity increases, however, the three-phase region expands more and the expanded three-phase region allows the bubble penetration to be easier. Thus, the gas holdups increase concavely with increasing gas velocity as shown in Fig. 2. In addition to this, because of the large density difference between particles and fluid, solid holdups show some fluctuations before full fluidization. On the other hand, when we compared Fig. 3 with Fig. 4, we could hardly find any big differences between those two figures. Thus, we measured the critical fluidization velocities of those two cases.

2. Critical Fluidization Velocity

Fig. 5 shows the change of critical fluidization velocity with respect to the change of particle loads. In general, the critical fluidization velocity decreases with increasing particle loads. This is a characteristic of three-phase inverse fluidized beds. Unlike the nor-

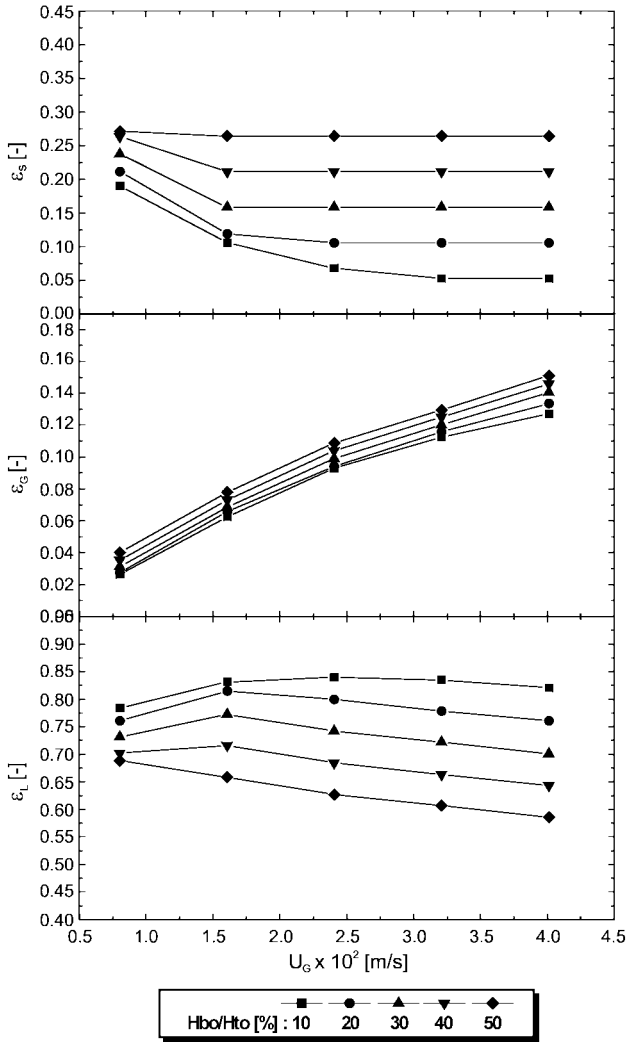

Fig. 4. Effect of gas inlet velocity on the individual phase holdups in three-phase inverse fluidized beds of MDPE hydrophobic particles.

mal fluidization of particles from bottom to top, larger loads are better for the fluidization of particles in case of inverse fluidization. When we compared three different cases, hydrophobic LDPE particles showed largest U_{GC} and hydrophobic MDPE showed small U_{GC}. There are two particle properties determining the critical fluidization velocity at the same size of fluidized bed: the density of particles and the surface property of particles. As we can easily expect, small density difference between particles and fluid allows easy fluidization. As shown in Fig. 5, the critical fluidization velocity is more sensitive to the density of particles than the surface property of particles. If the density of the particles is the same, however, hydrophobic particles show smaller U_{GC} at the same particle load than hydrophilic particles. To explain this phenomenon, we analyzed our experimental data in the viewpoint of the rising velocity of bubbles in the following sub-section.

We also compared our experimental data with other previous studies. As shown in Fig. 5, Comte et al. [1997] and Choi et al. [1999] also reported similar results. Comte et al. [1997] used PE particles with the density of 934 kg/m^3 in the fluidized beds equipped with both perforated plate and membrane distributors. Their data are located in the density between 926 and 940 kg/m^3. Although they used the same density of particles, the data show different critical velocities. This is due to the use of different distributors. Since a membrane distributor normally generates small bubbles, the U_{GC}s for a membrane distributor are smaller than those for a perforated plate distributor. Choi et al. [1999] used a small fluidized bed column with a height of 0.5 m equipped with a ceramic membrane distributor. As a result, the critical fluidization velocities became the smallest among other studies even at small density of particles. This must be the effect of bed size. Thus, the selection of distributor type and column size is also important in inverse fluidization.

3. Bubble Swarm Velocity

U_{GC} is the gas velocity when solid particles are uniformly distributed in the whole fluidized bed. Therefore, the decrease of U_{GC} with increasing particle loads means the decrease of gas inlet velocity and, of course, the bubble swarm velocity also strongly depends on the gas inlet velocity. As particle loads become large, the initial solid bed before fluidization occupies a large portion of the total bed and only a small injection of gas allows the average density of the bottom fluid phase to be lower than the density of the solid. This may be the reason for the decrease of U_{GC} with increasing particle loads. Thus, large particle loads result in the decrease of U_{GC} and the decrease of gas inlet velocity causes the decrease of bubble swarm velocity. At small particle loads, however, the increase of U_{GC} not only results in the formation of large bubbles due to fast gas inlet velocity but also allows the coalescence of rising bubbles due to large free volume between bubbles. The coalescence of bubbles was observed at small particle loads and the resulting large bubbles quickly rose in the fluidized bed. Thus, the bubble swarm velocity depends on both the gas inlet velocity and the particle loads.

Fig. 6 shows the variation of the bubble swarm velocity with respect to the change of particle loads. In case of hydrophobic LDPE particles, the bubble swarm velocity does not change much up to particle loads of 30%. This is because the resistance of rising bubbles becomes small due to low contact frequency with particles distributed in the bed under the condition of full fluidization and the gas inlet velocity is already too high for fully fluidizing particles with relatively small density (926 kg/m^3). When the loads become over 40%, however, the bubble swarm velocity quickly drops down due to the increase of frequency colliding with particles. But, high bubble swarm velocity mainly results from the large density difference between particles and fluid in this case. In cases of hydrophilic LDPE and hydrophobic MDPE, the bubble swarm velocity becomes small. This is mainly due to the small density difference between particles and fluid so that even small U_{GC} allows full fluidization of particles. While the bubble swarm velocity of hydrophobic MDPE particles decreases rather monotonically and becomes constant at high solid holdups, the bubble swarm velocity of hydrophilic LDPE particles decreases with showing some fluctuations and shows rather higher values than the former case. Since those two particles have the same density, this difference must come from different surface affinity with rising bubbles. Since the surface properties of both rising bubbles and falling MDPE particles are hydrophobic, the affinity between bubbles and particles slows down the bubble swarm velocity. On the other hand, the hydrophilic LDPE particle shows fluctuating character with increasing solid holdups.

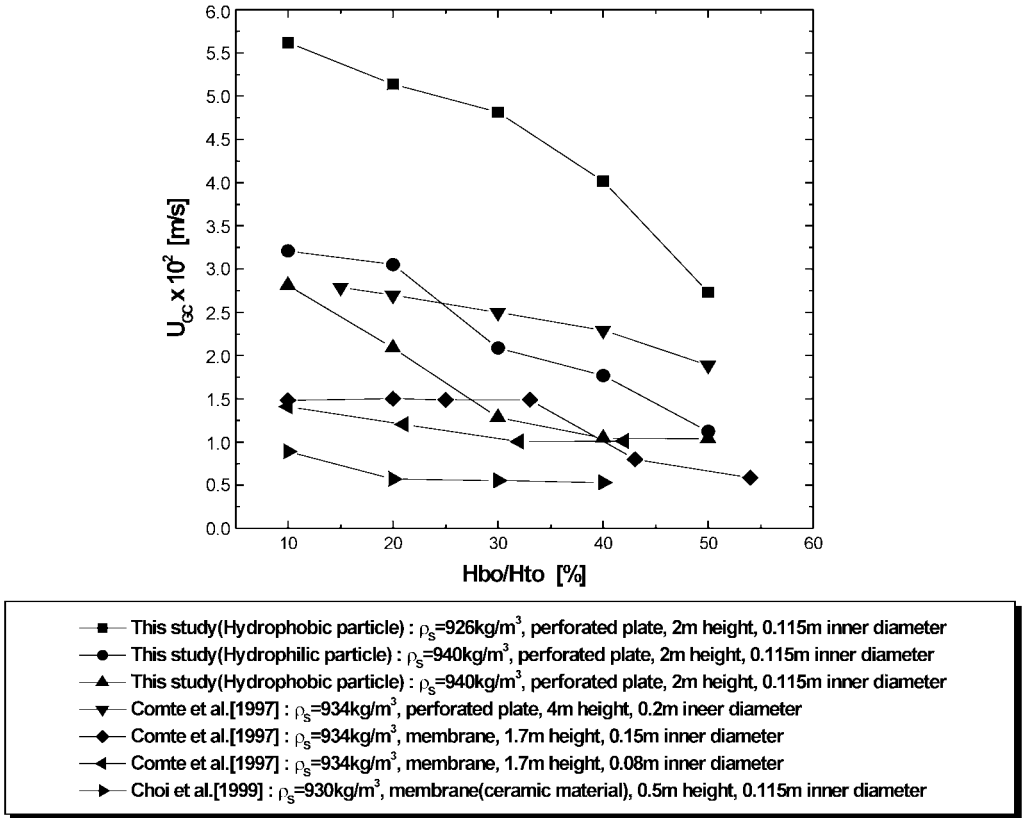

Fig. 5. Variation of the critical fluidization velocity with respect to particle loads.

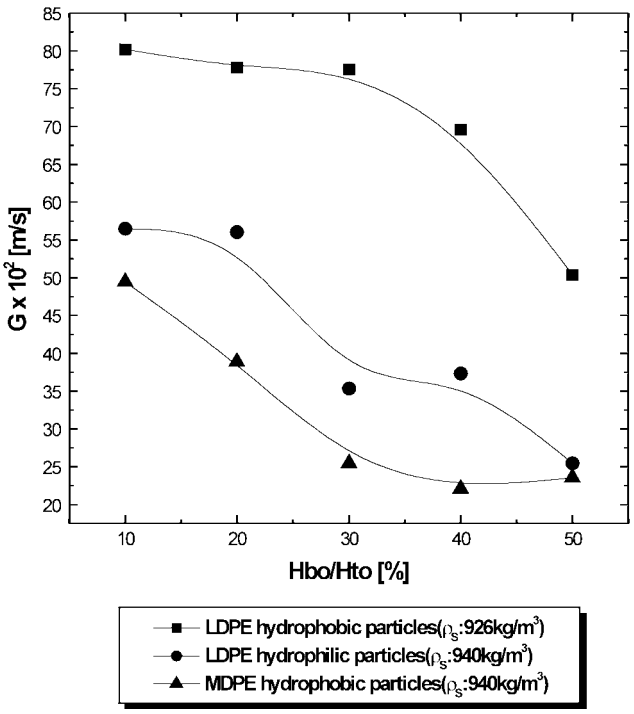

Fig. 6. Variations of the bubble swarm velocity with respect to the change of particle loads.

Finally, at high solid loading of H_{b0}/H_{t0}>20%, the bubble swarm velocities of two systems become almost the same due to the high blocking of rising bubble by particles.

CONCLUSION

In this study, we investigated the characteristics of phase holdups and critical fluidization velocity in three-phase inverse fluidization when we used three different particles of hydrophobic and hydrophilic LDPE particles, and hydrophobic MDPE particles with the same density as that of hydrophilic LDPE particles. First, we checked the effect of the density of particles on the hydrodynamic characteristics with particles having the same surface properties but different densities. Second, we investigated the effect of the surface properties on the hydrodynamic characteristics of particles having the same densities but different surface properties, which are hydrophobic and hydrophilic. In the three-phase inverse fluidization, the hydrodynamic characteristics such as phase holdups and critical fluidization velocity sensitively depend on the density difference of particles and fluid, and a small density difference makes inverse fluidization easier. When two particles have the same densities but different surface properties, hydrophobic particles are better for the inverse fluidization than hydrophilic particles in liquid water. Thus, when we operate the three-phase inverse fluidization, we should consider the surface properties of particles as well as the density of particles for easy inverse fluidization. In addition to this, the selection of gas distributor and column size is also very important to determine the critical fluidization velocity of inverse fluidization.

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NOMENCLATURE

A : cross section area of the column [cm^2]
$-dp/dz$: axial pressure gradient [$Pa \cdot m^{-1}$]
Dp : particle diameter [mm]
g : gravitational acceleration [$m \cdot s^{-2}$]
G : bubble swarm velocity [$cm \cdot sec^{-1}$]
G_0 : swarm velocity for $\varepsilon_S=0$ [$cm \cdot sec^{-1}$]
H_{b0} : bed height [cm]
H_{t0} : total bed height [cm]
M_s : weight of solid particles [g]
$R=1-\rho_s/\rho_L$: density ratio [-]
U_{GC} : critical fluidization velocity for gas [$cm \cdot sec^{-1}$]

Greek Letters

ε_G : gas holdup [-]
ε_L : liquid holdup [-]
ε_S : solid holdup [-]
ρ_G : gas density [$g \cdot cm^{-3}$]
ρ_L : liquid density [$g \cdot cm^{-3}$]
ρ_S : solid density [$g \cdot cm^{-3}$]

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