Formation of Particle Agglomerates after Switching Fluidizing Gases

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When a fluidizing gas is switched from a low-density gas to a high-density gas, particles in the emulsion phase agglomerate and bubbles disappear. At the same time, channeling occurs and a decrease in the pressure drop over the bed is observed. The disturbance of fluidization is temporary, and normal fluidization is restored after several minutes. The study of mechanisms of the transient phenomena shows that the difference in diffusion rates of the two gases in the emulsion phase caused the agglomeration of particles. Because the mobility of particles was reduced, channels were formed and the fluidizing gas bypassed through them. Effects of the properties of the fluidizing gases and particles on the intensity of the transient phenomena were also studied. The intensity increased with increasing difference in the densities of the two gases. The transient phenomena were considerable for small- and light-particle systems.

Introduction

During the operation of fluidized beds, switching of the fluidized gas is often carried out. For example, an inert gas is switched to a reactant gas during operational startup. When the fluidizing gas is suddenly changed from a low-density gas to a high-density gas, the emulsion phase is compressed and the bubbles disappear. At the same time, channeling and a decrease in the pressure drop over the bed are observed. The disturbance of fluidization is temporary, and normal fluidization is restored after several minutes. On the other hand, this phenomena is not observed when the gas is switched in the reverse direction, from a high-density gas to a low-density gas. Figure 1 illustrates this phenomenon as seen in a two-dimensional fluidized bed. The dark area corresponds to the emulsion phase. Figure 1a shows the bubbling fluidization when the bed was fluidized by helium gas, and good fluidization was observed. Figure 1b shows the fluidizing state at 20 s after switching the gas from helium to argon gas. Bubbles disappeared, the particles agglomerated, and channeling occurred. Figure 1c shows the fluidizing state at 40 s after the switching. The particle agglomeration was partially broken near the wall of the channels. The poor fluidization continued for about 80 s, and the fluidization quality was restored after 120 s in this case.

This phenomenon was originally reported by Rietema and Hoebink (1977). They experimentally studied the effect of the gas properties on the intensity of the phenomena during the transient period and attempted a theoretical analysis. They stated that this disturbance was unrelated to the gas adsorption on the particles because the transient phenomena were observed for both porous catalyst particles and glass beads.

Two mechanisms were proposed by Rietema and Hoebink. The first mechanism resulted from the difference in the emulsion-phase voidage, which was influenced by the viscosity of the fluidizing gas. The emulsion-phase voidage increases with the viscosity of the fluidizing gas for Geldart's A group particles (1973) as correlated by Abrahamsen and Geldart (1980) and Kai et al. (1987). The second mechanism resulted from the compression of the emulsion phase and bypassing of the fluidizing gas. It was explained by Rietema and Hoebink (1977) that the compression was caused by the local pressure gradient that was brought about by a net mass flow due to the difference in the densities of both gases.

They correlated the intensity of the transient phenomena with the gas properties based on the proposed mechanisms. However, because the correlation was obtained from limited experimental data, it is difficult to judge which mechanism is dominant. In addition, the process of generation of the pressure gradient by the net mass flow is not clear in the second mechanism. The objective of the present study is to investigate the mechanism of the transient phenomena. The effects of gas velocity, gas properties, and particle properties on the intensity of the phenomena were also investigated.

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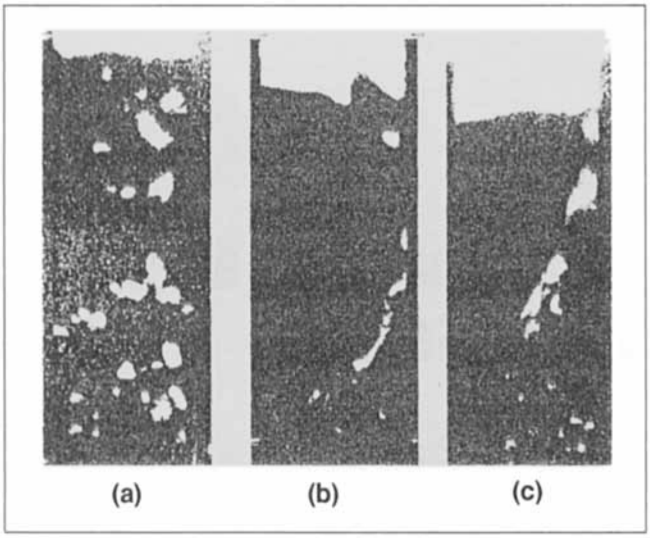

Figure 1. Channeling occurrence on gas switching from helium to argon in a 2-D bed: (a) before gas switching; (b) after 20 s; (c) after 40 s.

Experimental Equipment and Procedure

Particles and gas

Eight kinds of gases from gas cylinders were used as fluidizing gases: hydrogen, helium, nitrogen, ethylene, argon, propylene, carbon dioxide, and propane. The properties of the particles are listed in Table 1. The particles categorized in Geldart's A group (Geldart, 1973) were mainly used in the present study. The surface areas of the particles are also listed in Table 1. These were measured by the nitrogen-adsorption method in which the BET equation was used for calculation.

Fluidized bed

The fluidization column was made of transparent acrylic resin pipe with an inner diameter of 0.055 m and a height of 2.0 m. The gas distributor used was a sintered metal plate. Pressures in the bottom of the bed and in the freeboard region were measured using pressure transducers (P-A300, Copal Electronics Co., Ltd.). After the analog signals from the transducers were converted to digital signals, they were processed using a computer.

Table 1. Properties of Particles

Particles	d_p μm	ρ_p kg·m^{-3}	$S_a \times 10^{-3}$ m^2·kg^{-1}
Alumina			
Al-1	55.0	684	247
Al-2	75.2	780	141
Silica – alumina			
FCC-1	35.8	976	584
FCC-2	37.4	1,170	434
FCC-3	34.7	1,740	150
FCC-4	48.4	1,740	116
FCC-5	56.3	1,036	757
Silica			
Si-1	69.3	641	7.3
Si-2	103	1,360	387
Catalyst			
Cat-1	59.3	1,230	305
Cat-2	49.0	1,040	283
Cat-3	40.1	2,240	71
Cat-4	51.0	2,330	170
Glass beads			
GB	72.7	2,460	< 1

Before switching the fluidizing gas, the first gas was supplied to the bed for 600 s. The solenoid valve connected to the line of the second gas was opened the moment the first solenoid valve was closed. The actions of these valves were controlled by the signals from the computer. The values of the pressure drop during these operations were measured and recorded by the computer.

Packed bed

We investigated the relationship between the pressure change at the bottom of a packed bed and the diffusion rates of the gases in the packed bed. The experimental method was fundamentally the same as that reported by Rietema and Hoebink (1977). The experimental apparatus in the present study is shown in Figure 2. The inner diameter of the column was 0.02 m. The bed was filled with gas-a by feeding it from the bottom of the bed. The exit of the column was open to the atmosphere; therefore, the pressure at the bottom of the bed gradually reached atmospheric pressure after valve-1 was closed and the flow of gas-a was stopped. Gas-b was then supplied into the column just above the bed surface. Gas-b diffused into the bed and gas-a was exchanged with gas-b. The change in the pressure at the bottom of the bed was measured using a pressure transducer and a U-tube manometer.

Results

Figure 3 shows a typical curve of the pressure-drop decrease in a fluidized bed. In this case, the fluidizing gas was changed from helium to argon. When the gas flow was changed in the reverse direction, this change in the pressure drop was not observed. When the bed was fluidized by helium, the pressure drop was about 1,900 Pa. It decreased to about 1,200 Pa after the switch from helium to argon. At this time, agglomeration of particles and channeling were ob-

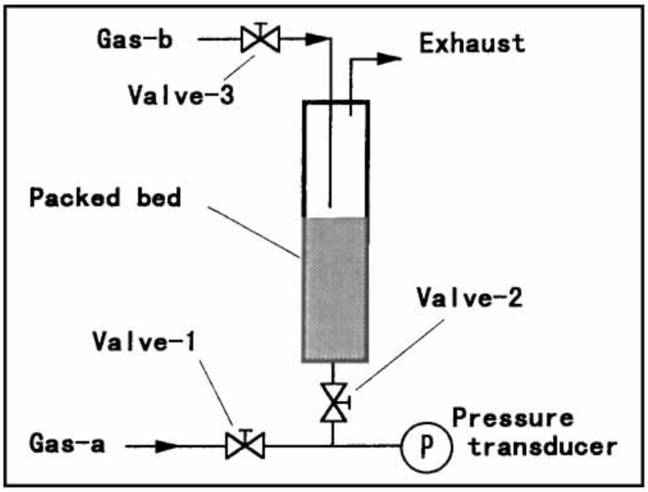

Figure 2. Experimental setup of the packed bed.

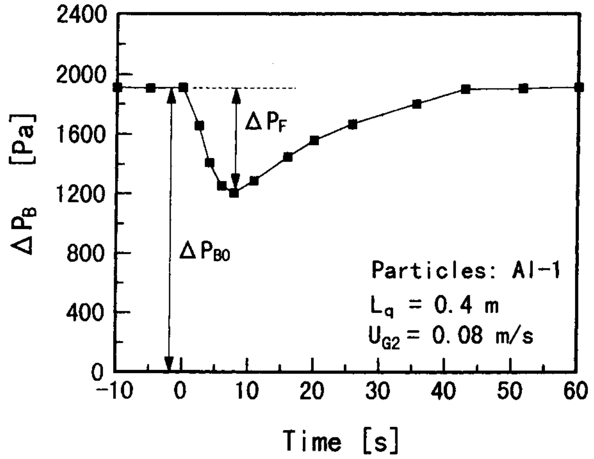

Figure 3. Change in pressure drop in the fluidized bed as a function of time after gas switching from helium to argon.

served and the bubbles disappeared. It took about 40 s for restoration to occur from the poor fluidization state. The intensity and the span of the transient period depended on the combination of fluidizing gases, gas velocity, and the properties of the particles.

Figure 4 shows the effects of the initial bed height of a fluidized bed on the intensity of the pressure drop decrease. As shown in Figure 3, ΔP_F is the maximum bed pressure-drop decrease during the transient period, and ΔP_{B0} is the pressure drop across the bed before gas switching. The absolute value of ΔP_F was affected by the bed height, but the relative intensity, $\delta = \Delta P_F / \Delta P_{B0}$, was not so affected by the bed height, as shown in Figure 4. During the transient phenomena, the bubbles disappeared and the mobility of particles in the emulsion phase was reduced. Although the pressure-drop measurement is a primitive method, it is considered suitable for evaluating the intensity of the phenomena. In the present study we therefore used the relative intensity, δ, which was obtained from the pressure drop.

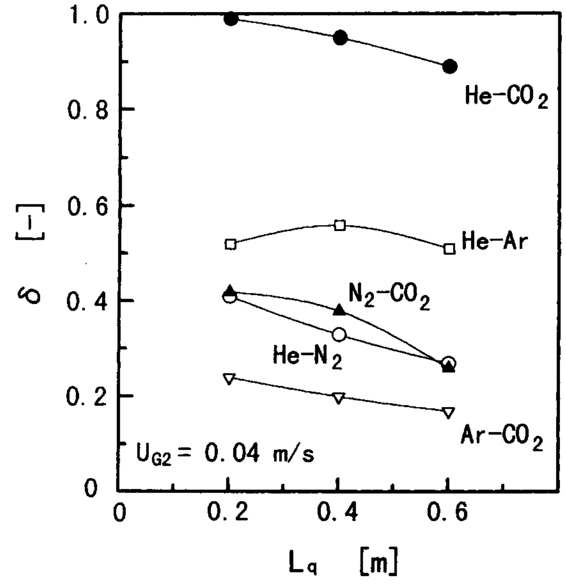

Figure 4. Effects of bed height on the relative intensity of pressure-drop decrease in the fluidized bed with Al-1 particles.

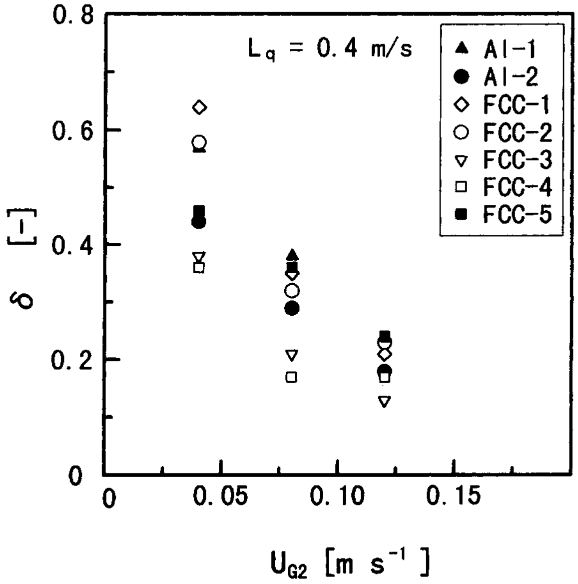

Figure 5. Effects of gas velocity on the relative intensity of pressure-drop decrease in the fluidized bed when switching the fluidizing gas from helium to argon.

Because the velocity of the fluidizing gas before the switching did not affect the intensity, the gas velocity before gas switching was fixed at 0.08 m s^{-1}. On the other hand, the gas velocity after the switching significantly affected the decrease in the pressure drop, as shown in Figure 5. A similar tendency was observed in all the particle systems. The overall volumetric gas exchange rate generally increases with the contact area between the emulsion phase and the gas phase, and with the extent of turbulence near the walls of channels. When gas velocity was raised, therefore, the rate of restoration was enhanced, and the intensity of the pressure-drop decrease was reduced. The span of the transient period also decreased with increasing gas velocity.

The relative intensity was conveniently plotted as a function of $d_p \cdot \rho_p$ in Figure 6. The relative intensity decreased with the increasing value of this parameter. When the value of this parameter was larger than 0.14 kg m^{-2}, the transient phenomena were not observed. Based on this result, it is suggested that the transient phenomena occur only for the particles categorized as Geldart's A group, and especially for the particles that expand in the emulsion phase of the bubbling bed. Although we also investigated the effect of the surface area of the particles on the relative intensity of the pressure-drop decrease, there was no correlation between them. Therefore, the adsorption effect of the fluidizing gas was insignificant or small in the system used in the present study.

It was reported by Rietema and Hoebink that the relative intensity was significantly affected by the difference between the gas densities. As shown in Figure 7, it was found that the intensity was correlated with a function of molecular weight, $1/M_b^{1/2} - 1/M_a^{1/2}$. We will discuss in the next section why we used such a function.

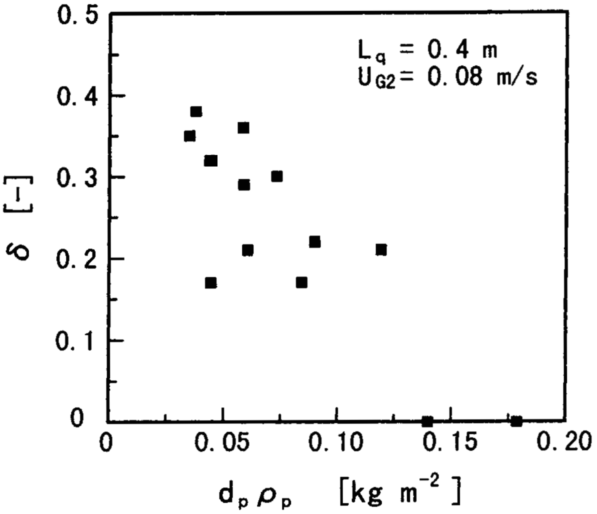

Figure 6. Effects of particle size and density on the relative intensity of pressure-drop decrease in the fluidized bed when switching the fluidizing gas from helium to argon.

Discussion

One mechanism suggested by Rietema and Hoebink (1977) was attributed to the difference in the emulsion-phase voidage due to the difference in gas viscosity. The emulsion-phase voidage generally increases with the viscosity of the fluidizing gas (Abrahamsen and Geldart, 1980; Kai et al., 1987). Rietema and Hoebink stated that a situation arose that appeared to be unstable when a fluidizing gas of low viscosity was replaced by another of high viscosity. Under this condition, the channeling that accidentally arose would grow, and the pressure drop over the whole bed would decrease. In the case of a reverse transition, any channeling was immediately suppressed. As they pointed out, this phenomenon shows a resemblance to the fingering effect that occurs when a high-viscosity oil in a consolidated sand formation is replaced by a low-viscosity fluid. However, they did not explain the reverse effect of viscosity in the case of the fluidized bed. The apparent bed viscosity decreased with increasing viscosity of the fluidizing gas because of an increase in the emulsion-phase voidage (Kai et al., 1991). Using the apparent viscosity of the bed, therefore it was found that the viscosity effect in the fluidized bed proposed by Rietema and Hoebink is similar to the fingering effect.

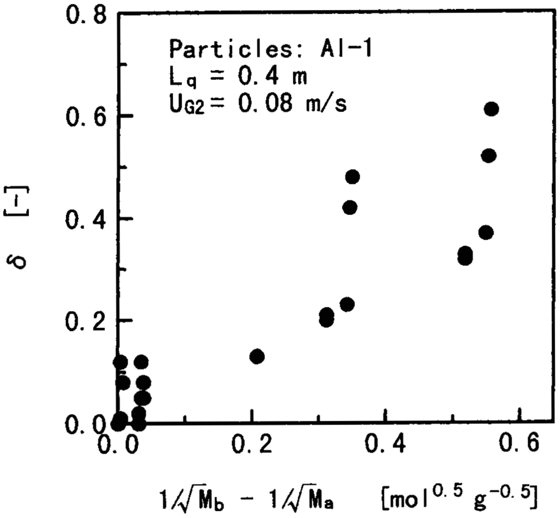

Figure 7. Effects of the difference between the molecular weights of the fluidizing gases before and after gas switching.

In the present study, we investigated the effect of gas viscosity on the transient phenomena in the fluidized bed with FCC-1 particles. We studied the systems of ethylene–nitrogen and carbon dioxide–propane. In each system, the molecular weights of the two gases are the same. Although FCC-1 particles exhibited a large decrease in the pressure drop after switching from a low-density gas to a high-density gas, the apparent decrease in pressure drop was not observed in the ethylene–nitrogen and carbon dioxide–propane systems. Consequently, the effect of the difference in gas viscosity is considered insignificant.

The other mechanism suggested by Rietema and Hoebink (1977) is attributed to a diffusion process due to high concentration gradients. They stated that the equimolecular diffusion process, which resulted from the concentration gradient, was accompanied by a net mass flow if the molecular weights of both gases were different. The emulsion phase was compressed because of the local pressure differences that arise as a result of the net mass flow. Large particle conglomerates formed because the mobility of the particles in these regions was decreased.

We investigated the pressure change in a packed bed to confirm this process. The pressure at the bottom of the packed bed was measured when the second gas was introduced just above the bed surface. When the bed initially contained a low-molecular weight gas, the pressure decreased after a high-molecular-weight gas was introduced. When the bed initially contained a high-molecular-weight gas, the pressure increased. Because the column vent led to the atmosphere, the pressure finally returned to atmospheric pressure. Figure 8 shows the results for the helium–argon system. When the first gas was helium and the second gas was argon, the pressure decreased and reached a minimum after 100 s.

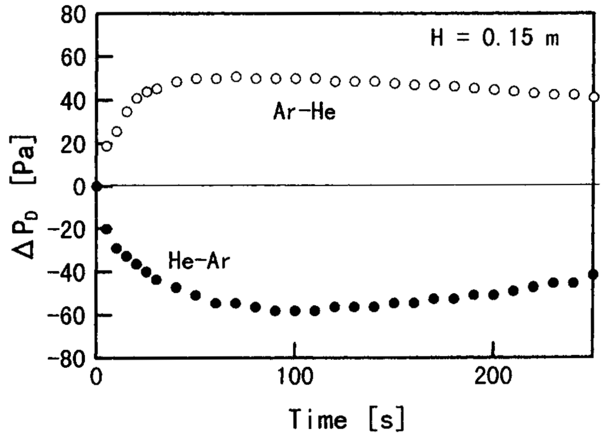

Figure 8. Pressure change with time in the packed bed for Al-1 particles.

In this case, it took about 1 h to return to atmospheric pressure.

If the pressure change is caused by the mass flow, the pressure change at the bottom of the packed bed should return to zero immediately when valve-1 and valve-2 in Figure 2 are closed. In the present study, however, the valve of the pressure at the bottom was maintained when these valves were closed. This result indicates that the gas concentration at the bottom part of the packed bed actually changed. This change cannot be explained by assuming equimolecular diffusion in the packed bed.

If the rates of the diffusion caused by the molecular movement of the two species are not identical in a binary mixture, there will be a concentration gradient. To maintain the requirements of an isobaric, isothermal system, bulk motion of the mixture occurs (Reid et al., 1984). Thus, equimolecular diffusion results both from diffusion and bulk flow. In a packed bed, however, the pressure gradient was not immediately canceled because of the presence of particle structures. Consequently, a pressure decrease or increase will be observed in the packed bed. We concluded that the extent of the pressure change in the packed bed was influenced by the difference in the diffusion rates of the two species.

Because the molecular velocity depends on the reciprocal number of the square root of the molecular weight, we assumed that the net molecular flow due to the difference in molecular velocities was presented approximately by the following equation:

$$N_D = -\alpha\left(\frac{1}{M_b^{1/2}} - \frac{1}{M_a^{1/2}}\right), \tag{1}$$

where α is a constant. When the molecular weight of the two components is different, the pressure gradient occurs in a packed bed and bulk motion of the mixture is caused by this pressure gradient. The molar flux due to the pressure gradient in a packed bed is given by the relation in the laminar flow region:

$$N_B = \frac{\beta \Delta P d_p^2 f(\epsilon)}{H\mu}, \tag{2}$$

where β is a constant, H the bed length, μ the viscosity of gas in the bed, and $f(\epsilon)$ a function of voidage. When the pressure in the bottom of the packed bed reached a maximum or minimum value, $-N_D$ balanced with N_B, the pressure change, ΔP_D is then given by:

$$\Delta P_D = \frac{\alpha H \mu}{\beta d_p^2 f(\epsilon)}\left(\frac{1}{M_b^{1/2}} - \frac{1}{M_a^{1/2}}\right). \tag{3}$$

Figure 9 shows the relation between the bed length and ΔP_D for the helium–argon system with Al-1 particles. The pressure change increased almost linearly with the bed length. Figure 10 shows the relation between particle diameter and ΔP_D. In this case, ΔP_D was measured in the helium–argon system at constant bed length. The pressure change increased with decreasing particle diameter and was almost proportional to d_p^{-2}. We investigated the effect of particle density on ΔP_D, but ΔP_D was found to be independent of particle density. A parameter concerning gas properties in Eq. 3 is presented by $\mu(1/M_b^{1/2} - 1/M_a^{1/2})$. Figure 11 shows the relation between this parameter and ΔP_D. Because the packed bed was filled with gas-a in the initial stage of the diffusion process, we used the value of the viscosity of gas-a for the calculation. Good correlation was obtained between the parameter and ΔP_D. These results correspond to the relationship presented by Eq. 3.

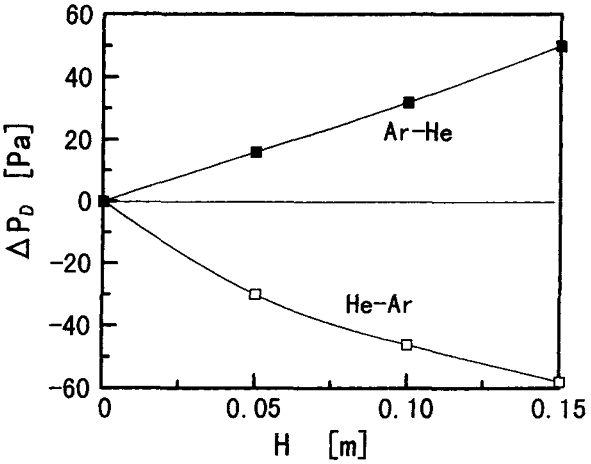

Figure 9. Effects of bed length of the packed bed on the pressure change for Al-1 particles.

We concluded from the preceding results that the pressure change observed in the packed-bed experiment was caused by the difference in the diffusion rates of the two gases. The same phenomenon is shown to arise in the emulsion phase of a fluidized bed after the fluidizing gas was switched. When the gas supply was changed from a low-density gas to a high-density gas, the gas in the bubble phase was replaced by a high-density gas within a few seconds, but the first supplied gas remained in the emulsion phase. Consequently, the emul-

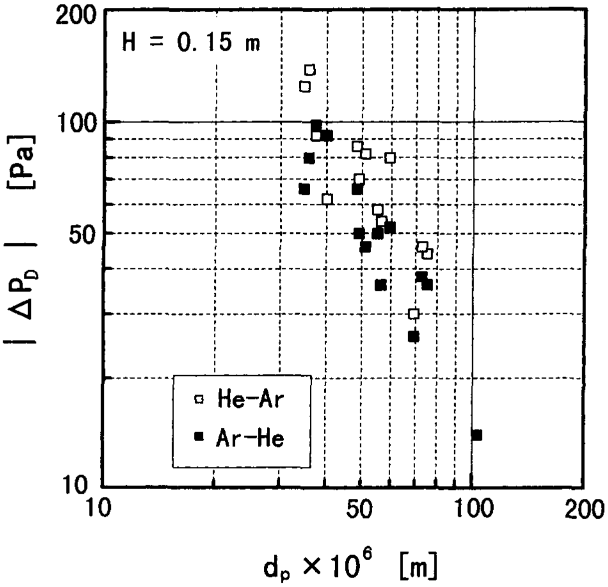

Figure 10. Effects of particle diameter on pressure change in the packed bed.

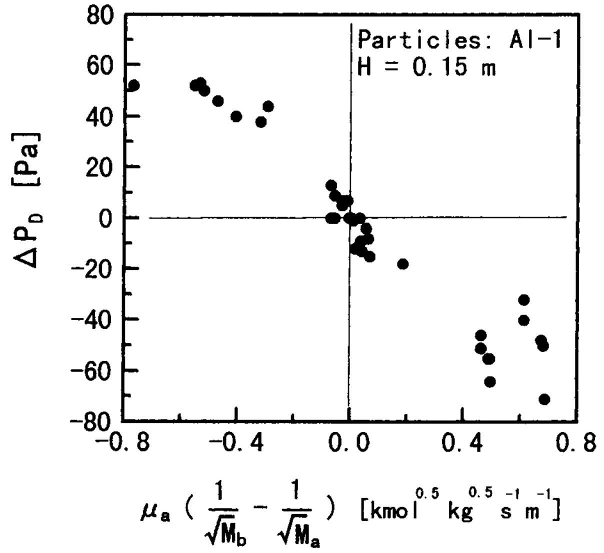

Figure 11. Correlation of the pressure change in the packed bed with a parameter containing gas properties.

sion phase was reduced and particle conglomerates were formed, as illustrated in Figure 12. Due to these phenomena, the fluidizing gas tended to bypass through the channels. On the other hand, when the fluidizing gas was changed in the reverse direction, the diffusion of the low-density gas to the emulsion phase was increased. As a result, the expansion of the emulsion phase was maintained and the decrease in the pressure drop did not arise.

The pressure change in the packed bed was influenced by particle diameter, and it was independent of particle density as described earlier. As shown in Figure 3, however, particle density also affects the pressure drop decrease in the case of the fluidized bed. The particle diameter of GB and Si-1 particles was about 70 μm, and the pressure change in the packed bed was almost the same. The pressure drop decrease was not observed in the fluidized bed with GB particles, whereas a significant decrease was observed for the bed with Si-1 particles. Because the gas velocity in the emulsion phase was large for heavy-density particles, the gas in the emulsion phase was exchanged quickly and the system was insensitive to the change in the gas properties.

The phenomena described in the present study occur not only when the fluidizing gas is switched, but the fluidization quality would, under reaction conditions, also be affected by the phenomena if molecules with small weight were produced by the reaction. The average molecular weight in the emulsion phase becomes smaller than that in the bubble phase in a reaction such as decomposition or disproportionation. In these reaction systems, the fluidization quality will not be as good as that predicted from the observation in a cold-model experiment.

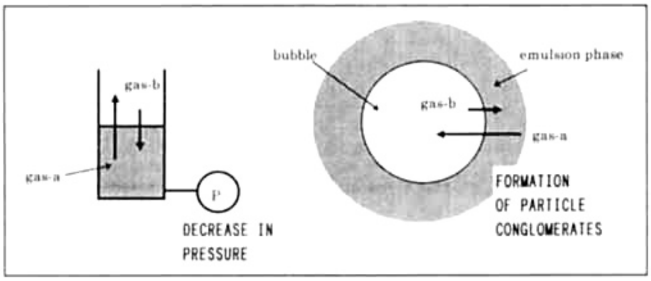

Figure 12. Molecular flow near the surface between gas phase and particle phase when the molecular weight of gas-a is smaller than that of gas-b: (a) packed bed; (b) fluidized bed.

Conclusions

When the fluidizing gas was switched from a low-density gas to a high-density gas, particle agglomeration and channeling occurred, and the pressure drop over the bed decreased. It was found that these phenomena were limited to Geldart's A group particles. The intensity of the phenomena increased with decreasing particle density and size. The influence of the difference in gas viscosity before and after switching is insignificant. However, the difference in gas density affects the fluidization quality when the fluidizing gas is switched. The particle agglomeration is considered to be caused by the difference in the diffusion rate of the two gases in the emulsion phase. This difference in the diffusion rate is due to the difference in the molecular velocity. To reduce the transient defluidization during gas switching, it is effective to temporarily increase the gas velocity after switching. If it is possible to select the gas type, choosing a gas system that minimizes the difference in gas density is recommended.

Acknowledgments

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Notation

d_p = particle diameter, m
L_q = settled bed height of the fluidized bed, m
M = molecular weight, $\mathrm{kmol \cdot kg^{-1}}$
N_B = molar flux due to bulk motion, $\mathrm{mol \cdot m^{-2} \cdot s^{-1}}$
N_D = molar flux due to the molecular movement, $\mathrm{mol \cdot m^{-2} \cdot s^{-1}}$
U_{G2} = superficial gas velocity after gas switching, $\mathrm{m \cdot s^{-1}}$
ΔP_B = pressure drop across the fluidized bed, Pa
ΔP_D = pressure drop due to diffusion in the packed bed, Pa
S_a = surface area of particles, $\mathrm{m^2 \cdot kg^{-1}}$
ρ_p = particle density, $\mathrm{kg \cdot m^{-3}}$

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