Effects of Increase in Gas Volume on the Fluidization Properties in Fluidized Bed Reactors

Takami KAI1, Jumpei HORINOUCHI1, Tsutomu NAKAZATO1, Toshio TSUTSUI1, Takeshige TAKAHASHI1 and Mitsuyuki NAKAJIMA2

1Department of Applied Chemistry and Chemical Engineering, Kagoshima University, 1-21-40, Korimoto, Kagoshima-shi, Kagoshima 890-0065, Japan
2Ishikawajima Plant Engineering & Construction Co., Ltd., 3-3-3, Harumi, Chuo-ku, Tokyo 104-0053, Japan

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There are many industrially useful reactions accompanied by the change of the gas volume due to the stoichiometry. Although it is important to consider the effects of the gas-volume changes for the accurate estimation of the reactor performance, the effects on the fluidization properties have not been experimentally studied. In this study, experimental simulation was carried out to study the effects of the increase in gas volume on the fluidization properties. The gas volume was increased by the evaporation of water impregnated in the pores of alumina particles at elevated temperature. The effects on the expansion of the emulsion phase, average bubble holdup, bubble size and pressure fluctuations were studied. The bubble growth, splitting and coalescence would become vigorous, whereas the emulsion phase expansion was not affected and the bubble size was slightly increased.

Introduction

Fluidized catalyst beds have been applied to many industrial catalytic reaction processes for around 60 years. Usually, reactions are accompanied by the change in the total number of moles due to the stoichiometry. In this case, the gas-volume changes (expands or reduces) under constant pressure and temperature. The volume of gas mixture increases in the case of the catalytic cracking of hydrocarbons, while it generally decreases in the case of hydrogenation. These changes lead to the vertical change in the gas velocity. The residence time differs from the calculation based on the gas velocity at the reactor inlet. The bubble size should be also influenced by the changed gas velocity.

The validity of proposed reactor models was intensively studied by analyzing reactions in fluidized catalyst beds in the early years of the fluidization research. Although these reactions often involved the gas-volume changes, many reactor models did not take account of the gas-volume changes. On the other hand, some researchers have emphasized that it is important to consider the changes in the total number of moles due to reactions in the fluidized bed reactor-models (Irani et al., 1980; Corella and Bilbao, 1984; Kai and Furusaki, 1987; Kai et al., 2004). The assumptions are different among the reactor models previously proposed. In order to satisfy the overall material balance at a given bed height, the crossflow terms have been considered in the reactor models (Kai and Furusaki, 1984; Shiau and Lin, 1993; Tafreshi et al., 2000). Irani et al. (1980) did not consider the crossflow, because they assumed that the extra gas generated went to form new bubbles. In addition, they considered that the effective diameter of the bubbles remained constant. On the other hand, the bubble diameter was assumed to change according to the gas velocity with height by some researchers (Corella and Bilbao, 1984; Shiau and Lin, 1993; Al-Zahrani et al., 2001).

In addition to the change in the residence time, the volume change leads to significant change in the hydrodynamic behavior of the bed. It has been reported that the fluidization quality was drastically decreased when the reactions involving a decrease in the gas volume were carried out in a fluidized catalyst bed (Kai and Furusaki, 1987; Kai et al., 2004).

In normal fluidized beds, a part of fluidizing gas flows through the emulsion phase. The effective bed weight is balanced by the gas drag force in the emulsion phase. In the case of the fluidized catalyst beds with Geldart’s A particles, the gas velocity is rather higher than the minimum fluidizing gas velocity (Abrahamsen and Geldart, 1980). The gas velocity in the emulsion phase will decrease when the reaction involving a decrease in
gas volume is carried out at constant temperature. When the degree of the gas-volume decrease is large, the gas supply from bubbles cannot compensate the volume decrease in the emulsion phase. In this case, the gas drag force becomes small not enough to balance with gravitational force and buoyancy force acting on particles in the emulsion phase. Consequently, the voidage of the emulsion phase reduces and the decrease in the fluidity of the emulsion phase disturbs bubbles to rise. When the rate of the volume decrease was large, defluidization in the bed occurred and the defluidized part was lifted up like a moving piston by the reactant gas (Kai et al., 2006). The reactor cannot be safely operated at these conditions.

On the other hand, the gas-volume increase due to reactions acts to increase the gas velocity and voidage in the emulsion phase. Therefore, at this condition the fluidization quality will be improved. Although many useful industrial reactions involve an increase in the gas volume, the influence of this change on the fluidization properties have not been investigated experimentally.

In the present study, the gas volume was increased by experimental simulation instead of reactions. The simulation was carried out in a fluidized bed at elevated temperature by the evaporation of water impregnated in the pores of alumina particles. The effects of the gas-volume increase on expansion of the emulsion phase, bubble holdup and bubble size were investigated.

1. Experimental

Porous alumina particles were used as the fluidizing particles. Distillated water was impregnated to the pores of the particles. When the particles are fluidized at an elevated temperature, the volume of fluidizing gas is expanded by the evaporation of water kept in the pores. The rate of the gas-volume increase was controlled by the bed temperature. Since the water content in the pores was limited, the steady state continued only for several minutes. However, it was enough for measurements.

The column made of glass with a 50 mm inner diameter was used as the column of the fluidized bed. A transparent electrical resistant material was coated on the outer surface of the glass tube and it worked as an electrical heater. When the column was heated, the transparency was sufficient to observe the inside of the column. The settled bed height was measured after the bed was fluidized and settled. The value was around 0.35 m. Since the bed height did not change below \(U_{mf}\), we used the settled bed height as the bed height at \(U_{mf}\). The superficial gas velocity at the inlet was varied from 0.015 to 0.06 m s\(^{-1}\). The superficial gas velocity at the outlet was calculated by assuming that the gas was saturated by water vapor at the bed temperature. The validity of this assumption remains to be discussed.

The average size of dried particles was 55 \(\mu\)m, and the particle density was 770 kg m\(^{-3}\). The minimum fluidizing velocity of the dry particles measured by air under ambient conditions was 1.9 mm s\(^{-1}\). Distillated water was impregnated to the pores of particles by spraying with the water over the particles. The particles were mixed during the operation. The load of the water in the particles was below 18% to keep a good fluidization condition. After fluidization was started with air at ambient temperature, the bed temperature was increased at 0.2 K s\(^{-1}\) and was finally controlled at prescribed temperature.

The expansion of the emulsion phase was measured by the bed collapse method (Rietema, 1967) when the bed temperature and the evaporation rate reached the steady state. The bubble holdup was obtained through the measurements of the settled bed height and the fluidized bed height.

The average bubble frequency was measured by inserting an optical probe. A crystal rod with 2 mm outer diameter was vertically inserted into the bed from the top of the column. Light was supplied from the light source to this rod by means of an optical fiber. When the tip of the rod was surrounded in the emulsion phase, the reflected light was detected by a sensor. On the other hand, when the tip was involved in a bubble, emission was diffused in the bubble and the intensity of the light reflection was weak. In this way, the presence of a bubble could be detected.

The fluctuations of pressure drop were measured by digital pressure sensor. The pressure port was a stainless steel tube with 2 mm inner diameter. The tube was vertically inserted in the bed from the top of the column. The sampling frequency of the sensor was 285 s\(^{-1}\).

2. Results and Discussion

2.1 Change in the bed height

Since the column made of glass was used, its inside could be directly observed. Judging from the fluctuation of the bed height, slugging was not occurred under all the experimental conditions.

Figure 1 shows the change in the bed height when the temperature was increased from 300 to 363 K. The
left hand Y-axis is the relative bed height based on the bed height at 300 K. The initial water content of the particles was 17%. The temperature, the right hand Y-axis, reached 363 K after 350 s. The bed height began to increase immediately with a temperature increase. After the bed-height change reached the steady state, the gases at the outlet were saturated with water vapor at this temperature, and the expansion of the fluidizing gas could be considered to be stable. Measurements were carried out during this period. Since the most part of the water in the pore had evaporated during initial 600 s, the evaporation rate decreased and the bed height gradually dropped after 600 s. Since the temperature was elevated, the bed height did not return to the initial bed height after all the water had evaporated.

2.2 Emulsion phase expansion

The emulsion phase is expanded when fine particles are fluidized. The expansion ratio is influenced by the particle properties and gas properties (Abrahamsen and Geldart, 1980). The voidage of the emulsion phase, \( \varepsilon_{fe} \), is correlated by the correlation obtained by Kai et al. (1987):

\[
\varepsilon_{fe} = \varepsilon_{mf} + \frac{4.11 \mu p^{0.12}}{g d^3 (\rho_p - \rho)}
\]  

(1)

The relationship between \( \varepsilon_{fe} \) and \( \varepsilon_{mf} \) can be obtained based on the balance of solid:

\[
L_e (1 - \varepsilon_{fe}) = L_{mf} (1 - \varepsilon_{mf})
\]  

(2)

where \( L_e \) is the emulsion phase height measured by the bed collapse method and \( L_{mf} \) is the bed height of the settled bed. The expansion ratio, \( L_e / L_{mf} \), of the dry alumina particles measured at ambient temperature was 1.13. The expansion ratio was 1.11 when 9% of water was impregnated to the pores. The difference was due to the apparent particle density caused by water loading.

Figure 2 shows the bed expansion when the bed temperature was increased. The values of \( U_{Gout} / U_{Gin} \) were changed by the final temperature. All the values were around 1.1 and the expansion ratio was not so affected by the ratio of gas velocities.

The effects of the gas properties on the emulsion phase voidage were checked, since the mole fraction of water vapor increased with temperature. The expansion ratios were calculated by Eqs. (1) and (2) and compared for two cases. One is the case when the bed was fluidized by air at ambient temperature. The other is the case when the bed was fluidized by the air saturated with water vapor at 363 K. The calculation indicated that the difference between these values was only 2%. This result indicates that the emulsion phase voidage did not influenced by the increasing fraction of the water vapor under this condition.

Therefore, the results show that the expansion of the emulsion phase was not affected by gas-volume increase and was dominated by the gas properties and particle properties. The results also indicate that the generated extra gases immediately move to the bubble phase.

2.3 Bubble holdup

Average bubble holdup, \( \bar{\varepsilon}_b \), was obtained from the height of fluidized bed and emulsion phase height. The average bubble holdup increased with the ratio of gas velocities at the outlet and the inlet of the bed as shown in Figure 3. Although the initial water content affected the duration of the steady state, the average bubble holdup was not affected by the water content. These results justify the assumption that the outlet gas was saturated with water vapor at each temperature.

2.4 Bubble size

A part of the generated extra gases is absorbed by rising bubbles. The other gases form new bubbles. If the former phenomenon is dominant, the bubble size will be larger than that without the gas-volume increase. The bubble size was estimated by measuring the bubble frequency in the bed.
Figure 4 shows the relationship between the gas velocity at the outlet and the bubble frequency. The particles initially contained 12–16 wt% of water. The measurements were carried out at the center of upper part of the bed, the height of which was 0.3 m from the bottom. Since the measurement was carried out at a single point, all the bubbles that passed through this horizontal plane were not detected, that is, and the average bubble diameter could not be calculated correctly. In addition, since the bubble and emulsion phases were distinguished at a threshold value, a group of bubbles could not be correctly measured and a small bubble was missed. Consequently, the obtained bubble frequency was underestimated. For these disadvantages, the bubble frequency is still one of useful parameters to estimate the bubble size.

The open keys in this figure show the results when the temperature was 303 K. The gas volume did not increase at this temperature. When the bubble size is not affected by the gas velocity, the bubble frequency is proportional to $U_{G} - U_{mf}$. In the present study, however, the bubble frequency was lower than the values estimated by the liner relationship. Therefore, the average bubble size would increase with increasing gas velocity.

The values of the bubble frequency when the gas volume was increased were shown as the closed keys in this figure. The temperature was increased to 353 K and the value of $U_{Gout}/U_{Gin}$ was about 2. The bubble frequency was rather smaller and the bubble size was considered to be slightly larger than that corresponding to the velocity without gas-volume increase. For fine particles like the particles used in this study, bubble growth is not dominant in the upper part of the bed, and bubble splitting and coalescence reached an equilibrium (Miyauchi et al., 1981). In addition to the splitting and coalescence of bubbles, bubbles absorb the generated extra gases when the gas volume increases. The absorption of the gases would lead to the slight increase in the bubble size.

2.5 Pressure fluctuations

The average bubble diameter calculated based on the data in Figure 4 was almost same as the column diameter. Judging from the signal pattern of pressure fluctuations, however, the slugging was not detected under the experimental conditions. The bubble size could not be correctly determined from the bubble frequency measured in this study as described above.

Figure 5 shows examples of signals of pressure fluctuations. The initial gas velocity was 0.03 m s$^{-1}$ and temperature was increased to 353 K. Figure 5(a) shows the results when dry particles were used. When the wet particles were used and the gas-volume increase was accompanied, larger fluctuations were observed as shown in Figure 5(b).

The average deviation of amplitude of pressure fluctuations, $\delta_p$, was calculated from the measured data based on the following equation:

$$\delta_p = \left( \frac{1}{n} \sum P_i - \frac{1}{n} \sum P_i \right)^2 / \frac{1}{n} \sum P_i$$

(3)

Figure 6 shows the effects of the gas-volume increase on $\delta_p$. The amplitude of the fluctuations increased with the superficial gas velocity at the inlet. When the gas
volume increased in the fluidized bed, the amplitude of the fluctuations was significantly larger than that without gas-volume increase. When the superficial gas velocity at the inlet was 0.015 m s$^{-1}$, it was increased and was about 0.03 m s$^{-1}$ at the outlet. The amplitude at this condition was about twice that for dry particles without gas-volume increase at 0.03 m s$^{-1}$.

When the gas-volume increases in the fluidized bed, the extra gases generated in the emulsion phase move into the bubble phase. Therefore, the fluidization properties are considered to be different from those of the bed without gas-volume increase. For example, the mass transfer rate between the bubble and the emulsion phase will be enhanced by the bulk flow from the emulsion phase to bubbles.

This flow will change the hydrodynamic behavior in the fluidized bed accompanied by the gas-volume increase as observed in the amplitude of the pressure drop fluctuations.

Conclusions

The effects of the gas volume-increase on expansion of the emulsion phase, bubble holdup and bubble size were investigated. For this purpose, the simulation was carried out by the evaporation of water impregnated in the pores of alumina particles in a fluidized bed.

The emulsion phase expansion was not influenced by the gas-volume increase. The generated extra gas was considered to move to the bubble phase immediately. In addition to bubble splitting and coalescence, some part of generated extra gases would be absorbed by bubbles. This would result in the slight increase in the bubble size.

The gas-volume increase did not much change the static fluidization properties such as the emulsion-phase expansion and the bubble frequency. Judging from the signals of pressure-drop fluctuation, the dynamic hydrodynamic behavior such as the bubble growth, splitting and coalescence would be activated by the gas-volume increase.

**Fig. 6** Effect of gas-volume increase on the deviation of the amplitude of pressure drop fluctuations

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**Nomenclature**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Units</th>
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<tr>
<td>$\bar{\rho}$</td>
<td>particle diameter</td>
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<tr>
<td>$g$</td>
<td>gravitational acceleration</td>
<td>[m s$^{-2}$]</td>
</tr>
<tr>
<td>$L_e$</td>
<td>equivalent height of emulsion phase</td>
<td>[m]</td>
</tr>
<tr>
<td>$L_{mf}$</td>
<td>bed height at $U_{mf}$</td>
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<tr>
<td>$n$</td>
<td>number of sampled data</td>
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<td>$U_G$</td>
<td>superficial gas velocity</td>
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<td>$U_{Gin}$</td>
<td>superficial gas velocity at bed entrance</td>
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**Literature Cited**


