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# Visualization of defluidization phenomena caused by gas switching in a two-dimensional fluidized bed

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Abstract: Transient defluidization occurs when fluidizing gas is switched from a lower density gas to a higher density gas. The intensity of this phenomenon increases with decreasing particle size and with increasing difference between gas densities. In order to investigate the mechanism of this effect, images of the transient phenomenon were captured using a video camera and visualized using digital image conversion. Gray-scale pictures were obtained based on the transmittance of back light through the emulsion phase in a thin two-dimensional fluidized bed. The gray-scale pictures were transferred into pictures with a color gradation. The generation, expansion, and elimination of defluidization after switching of the fluidizing gas can clearly be seen by observing the reconstructed color movies.

## 1. Introduction

Defluidization is observed when the reactions accompanied by a decrease in the number of moles are performed in fluidized catalyst beds [1–5]. While methanation of CO<sub>2</sub> has been performed in these studies, Go et al. [6] reported that defluidization could also be observed in the oxidation stage of two-step oxychlorination reactions. In addition, Abba et al. [7] suggested that a volume reduction in a fluidized bed reactor would lead to a decrease in gas velocity with the possibility of defluidization; although they did not conduct the necessary experimental work. Recently, Li and Guenther [8] studied the effect of gas volume change accompanied by ozone decomposition, and the reverse reaction, on the flow hydrodynamics of fluidized bed reactors using DEM simulations.

The gas velocity in the emulsion phase decreases as the reactions progress. When the decrease in the gas velocity in the emulsion phase is not completely compensated for by the gas supply from the bubble phase, the gas drag force becomes too small to balance the gravitational and buoyancy forces acting on the particles. Consequently, the gas velocity in the emulsion phase decreases below the minimum fluidization velocity, and the particles begin to agglomerate ultimately leading to defluidization. The poor fluidization continues while reactions are performed.

When the fluidizing gas is switched from a lower density gas to a higher density gas, defluidization is caused similarly due to the decrease in gas velocity in the emulsion phase. In this case, defluidization is transient and fluidization is restored after several minutes. This phenomenon was first reported by Rietema and Hoebink [9], while Kai and Takahashi [10] proposed the mechanism for this phenomenon. Thereafter, further studies on this phenomenon have been also published [11–13]. When the fluidizing gas is switched from a lower density gas to a higher density gas, the gas in the bubble phase is changed from a lower density gas to a higher density gas after several seconds. On the other hand, since the gas velocity in the emulsion phase is much lower than that in the bubble phase, it takes several minutes to replace the whole gas in the emulsion phase. Therefore, the situation in which the gas densities are different in the bubble and emulsion phases is maintained for several minutes.

The pressure drop across the bed decreases when defluidization occurs. The intensity of the decrease in pressure drop could be correlated with the reciprocal of the square of the particle diameter, and the difference between the reciprocals of the square root of the molecular weight of each gas [10]. It was supposed from this relationship that defluidization was affected by the difference in the mobility of molecules and the viscous flow caused by the pressure difference in the emulsion phase. When the gas is changed from a lower density gas to a higher density gas, the mole flux from the emulsion phase to the bubble phase becomes greater than that in the reverse direction: namely, a net gas movement from the emulsion phase to bubble phase occurs in the fluidized bed. Consequently, agglomeration and defluidization are caused by the decrease in gas velocity in the emulsion phase. Reichhold et al. [13] established a model based on this concept and some assumptions and showed that the tendency of the duration of the defluidization could be predicted well with the model.

Gas adsorption is the other mechanism of transient defluidization caused by gas switching. When the adsorptivity of the second gas is higher than that of the first gas, the gas velocity in the emulsion phase decreases, and transient defluidization is caused [11–13]. It was experimentally shown that the gas flow rate at the outlet largely decreased when the fluidizing gas was switched from He to CO<sub>2</sub> [13]. This influence of gas adsorption was significant for porous particles. Kai et al. [11] added a term for the capacity of adsorption to the index previously proposed for evaluation of the intensity of the decrease in pressure drop [10].

In order to study the mechanism, we needed to investigate where the defluidization begins and how this region

expands in the bed. Although the unusual fluidization phenomena can be detected in a three-dimensional bed by the change in pressure drop, we cannot accurately know what is occurring in the bed. On the other hand, in the case of a conventional two-dimensional bed, we can observe the channeling formation in the bed. However, it is not possible to discuss the defluidization phenomena in detail, because the bed voidage distribution has not been measured.

It is important from an industrial perspective to investigate this phenomenon, because the gas switching is carried out when a reactor is started up and shut down. The objectives of this study are to develop a color imaging method for the estimation of the voidage distribution of the emulsion phase, and to observe visually the bed behavior after the fluidizing gas is switched from a lower density gas to a higher density gas. For this purpose, we performed measurements using a thin two-dimensional fluidized bed and processed colored movies based on digital image conversion.

## 2. Experimental

The experiments were performed in a two-dimensional fluidized bed with a cross-section of  $80 \times 2$  mm and a height of 0.6 m. The column was constructed from a transparent glass sheet with a thickness of 3.0 mm. Alumina balls with a diameter of 0.267 mm were packed at the bottom of the column to distribute the fluidizing gas.

Porous alumina particles with an average diameter of 56  $\mu\text{m}$  and a particle density of  $790 \text{ kg m}^{-3}$  were fluidized. These particles are categorized to Geldart's A group [14], in particular to Ikeda's AA (advantageous aeration) group [15]. They showed very good fluidization quality. The expansion ratio of the emulsion phase during fluidization was approximately 1.17 when the fluidizing gas was He. The settled bed height was approximately 0.2 m throughout the experiments.

Five cylinder gases were used as fluidizing gases:  $\text{H}_2$ , He,  $\text{N}_2$ , Ar, and  $\text{CO}_2$ . After the flow rate was measured and controlled, the fluidizing gas was supplied from the bottom of the column. Although the distance between two panels of the column was small, and 2 mm, this was equivalent to the sum of the diameters of 35 particles. Therefore, small bubbles and good fluidization were observed when the bed was fluidized under steady state.

The fluidized bed was illuminated with back lighting and the motion of the bed was recorded using a video camera from the front of the column. There are many previous studies in which bed behavior was studied based on transmitted light using a two-dimensional bed. Digital image analysis has been developed to study bubble properties [16–19]. Sun and Grace [20] determined the apparent concentration of particles inside bubbles, while Yue et al. [21] studied the emulsion phase voidage. In addition, voidage distribution around bubbles could be measured [22].

In this study, before the gas switching, the first gas was supplied to the bed and the gas in the column was filled with the first gas. The bed behavior before and after the gas switching were recorded using a video camera. The pressure drop across the bed was also measured using a pressure port at the bottom of the column. The analog signals from a pressure transducer were converted to digital signals using an A/D converter, and recorded using a computer.

The emulsion phase in the fluidized bed of fine particles expands and its voidage is greater than that at the settled bed. Since the distance between the panels was only 2 mm, the difference in the voidage affected the intensity of light transmittance and a gray-scale pattern was recorded at each picture. Gray-scale values were adjusted by changing the sensitivity of the video camera and a gray-scale of each pixel ranging between 0 and 255 was converted into a color in the *RGB* space using the following non-linear transformations:

$$(R, G, B) = (255, -510(I - I_S)/(I_S - I_E), 0) \quad \text{for } I_S < I < (I_S + I_E)/2 \quad (1)$$

$$(R, G, B) = (510(I - I_E)/(I_S - I_E), 255, 0) \quad \text{for } (I_S + I_E)/2 < I < I_E \quad (2)$$

$$(R, G, B) = (0, 255, -510(I - I_E)/(I_S - I_E)) \quad \text{for } I_E < I < (3I_E - I_S)/2 \quad (3)$$

$$(R, G, B) = (0, 510(I - I_V)/(3I_E - I_S - I_V), 255) \quad \text{for } (3I_E - I_S)/2 < I < I_V \quad (4)$$

where  $I_S$ ,  $I_E$  and  $I_V$  are the average gray-scale values corresponding to the light transmittance of the settled bed, of the emulsion phase and of the vacant column, respectively. Because a part of the incident light transmitted through the settled bed,  $I_S$  was not zero. The value of  $I_E$  was obtained from the transmission through the emulsion phase without bubbles after the fluidizing gas was shut off when the bed was fluidized by He gas. The values of  $I_S$ ,  $I_E$  and  $I_V$  were estimated to be 76, 119 and 242, respectively. Fig. 1 shows the relationship based on Eqs. (1) – (4) between the gray-scale values and colors.

The light source was not flat and homogeneous. In addition, the emulsion phase was not homogeneous when the bed was fluidized. As the result, the gray-scale values depend on the position in the image. However, the effect of the ununiformity on the obtained images was insignificant. According to the transformation rule, the bubble phase, the emulsion phase and the defluidized region were clearly represented by blue, yellowish green and red colors, respectively. Color movies were developed by processing this conversion for each still picture composing monochrome movies.

### 3. Results and discussion

#### 3.1 Digital image conversion

Fig. 2a shows the gray-scale picture of the bed at 15 s after switching the fluidizing gas from Ar to He. The decrease in the fluidization quality did not occur by this gas switching: from a higher density gas to a lower density gas. Fig. 2b shows the colored picture according to the gray-scale level according to the method described above. These pictures show that small bubbles were rising uniformly in the bed. The average size of the bubbles was obtained directly from the picture to be approximately 6 mm. The fluidization quality was extremely good even though the thin two-dimensional beds were used.

Fig. 3a is the gray-scale picture of the bed at 15 s after switching the fluidizing gas in the reverse direction. Fig. 3b is the colored version of Fig. 3a. The voidage of the red region was almost the same as that of the settled bed. Bubbles did not exist in this region and this region was no longer fluidized. When the gas was switched from a lower density gas to a higher density gas, defluidization occurred and channels were formed. In this study, we obtained colored movies for a series of the phenomena. This enabled us to simultaneously observe the change of voidage distribution in the emulsion phase, the behavior of bubbles rising in the bed and the formation of channels.

#### 3.2 Generation of defluidization

Fig. 4 shows the emulsion phase voidage after 2 s of the gas switching. The low voidage spots developing in the middle of the bed as well as at the bottom of the beds are observed in both the pictures. These spots fell down in the emulsion phase when bubbles rose around them, and were captured by the defluidized region at the bottom of the bed. The defluidized region in the initial stage expanded via this mechanism.

Yellow was the dominant color for the emulsion phase in Fig. 4b, and this means that the defluidized region and the region with the small voidage were larger for the H<sub>2</sub>/Ar system (Fig. 4b) than for the He/Ar system (Fig. 4a). These results are attributed for two reasons. One is that the voidage of the emulsion phase before the switching is smaller when the bed is fluidized by the gas with lower viscosity and density [23, 24]. As the emulsion phase voidage decreases, the apparent viscosity of the bed increases and consequently bubble size increases [25, 26]. Larger bubbles were actually observed when H<sub>2</sub> was used as a fluidizing gas, as observed from the comparison of Fig. 4b with Fig. 4a. The other reason is that the diffusion rate is larger for H<sub>2</sub> than for He due to its smaller molecular weight. Since the first gas in the emulsion phase moved faster to the bubble phase in the case of the H<sub>2</sub>/Ar system, the voidage of the emulsion phase decreased more rapidly.

Fig. 5 shows the time series of colored pictures of the bed after the gas switching from He to Ar (see also Video Data). The superficial gas velocity of the second gas was 0.042 m s<sup>-1</sup>. These pictures were taken from the different run shown in Fig. 4a, whereas the experimental conditions were the same.

Some low voidage spots were found at the bottom immediately after the gas switching. The agglomeration was first generated at the bottom of the bed as described above. Fig. 6 shows in detail the change in the bed voidage at the bottom region. The leftmost picture was cut from the image at 0.2 s after the gas switching. The time interval of the pictures was 0.1 s and the size of this area was 47 × 55 mm. It can be seen that the voidage contraction occurred in the internal area of the emulsion phase. Since the gas could be supplied from bubbles, the voidage did not decrease in the emulsion phase near the bubble phase. In addition, the first defluidized region was generated at the bottom of the bed just above a distributor.

### 3.3 Expansion of defluidized region

After 3 s, the defluidized region expanded and a channel was clearly seen through this region (Fig. 5). The defluidized region containing no bubbles expanded to the upper part of the bed. This area was approximately 30% of the whole bed after 10 s, and reached 60% after 20 s.

Once the defluidized region was formed at the bottom of the bed, gas was no longer supplied to the emulsion phase above. Because gas tends to flow in channels due to the smaller flow resistance. Therefore, the emulsion phase turned to the defluidized region from the bottom as the gas escaped from this region. The rising velocity of the front line of the defluidized region was ascertained from the pictures of the He/Ar system as approximately 14.5 mm s<sup>-1</sup>.

Two regions which have a different voidage appear after the gas supply is shut off and bubbles escape when the bed collapse technique [27] is used. The upper region is homogeneously fluidized and its bed surface falls slowly at a constant rate. The lower region is a settled bed and the interface between the upper and lower regions rises at a constant rate. Schematic pictures showing the change in the bed surface and the interface are given in many papers previously published [28–31]. Fig. 7 shows clearly the change in the two regions after the fluidizing gas was shut off when the bed was fluidized by He gas. The rising velocity of the interface obtained from these pictures was 16.0 mm s<sup>-1</sup>. This value is almost consistent with the rising velocity of the defluidized region in Fig. 5. In this way, the

fluidized region expanded all throughout the bed mainly due to the insufficient gas flow to the emulsion phase after the fluidized region was generated and channels were formed at the bottom of the bed.

When the defluidized region expanded upward, the collapse of the defluidized region began from the bottom. The surface of the defluidized region was eroded by the gas-flow containing particles. Finally, the fluidization quality was restored after approximately 60 s in this case. At this time, small bubbles were observed though the whole bed. Fig. 8 shows the effect of the superficial gas velocity after gas switching on the time for the recovery of the fluidization in the He/Ar system. The time for the recovery decreased slightly with increasing gas velocity. Since the surface of the defluidized region was eroded to restore fluidization, the effect was larger for a larger gas velocity.

### 3.4 Effect of gas adsorption on defluidization

Fig. 9 shows the time series of the colored pictures when the gas was switched from He to CO<sub>2</sub> (see also Video Data). In addition to the difference between gas densities, since the second gas, CO<sub>2</sub>, adsorbed on the porous alumina, the gas velocity in the emulsion phase decreased rapidly and to a large extent. This effect enhanced the intensity of the defluidization.

The voidage distribution at 1 s after the gas switching was similar to that after the gas switching from He to Ar. In addition, channels were clearly formed after 3 s. However, in the case of CO<sub>2</sub>, the defluidized region expanded very rapidly and the whole bed was defluidized after 10 s. The surface of the defluidized region was also eroded by the gas-flow containing particles and fluidization was restored from the lower part of the bed. The maximum ratio of the defluidized region was large, and it took approximately 90 s to recover the fluidization quality.

When the gas was switched to Ar, the front line of the defluidized region was horizontal and expanded upward. However, when the gas was switched to CO<sub>2</sub>, the front line of the defluidized region was vertical and expanded in the horizontal direction after 5 s, as shown in the pictures in Fig. 9. This is because the gas adsorption on the porous particles was significant for CO<sub>2</sub>, and CO<sub>2</sub> diffused horizontally to the emulsion phase from channels. The horizontal velocity of the front line obtained from the pictures was approximately 20 mm s<sup>-1</sup> in the first 2 s.

In this study, this velocity is compared with the gas diffusion rate calculated on the basis of a simple model in a packed bed. The overall material balance in a differential portion of the fixed bed is given by Eq. (5).

$$\varepsilon \frac{dC}{dt} = \left( \frac{\varepsilon}{\tau} \right) D_{AB} \frac{d^2C}{dx^2} \quad (5)$$

where  $\varepsilon$  is the bed voidage,  $\tau$  the tortuosity, and  $D_{AB}$  the gas diffusivity.

Fig. 10 shows the calculations obtained with the initial conditions,  $C_A = C_0$  and  $C_B = 0$  at  $t = 0$ , and with the boundary condition,  $C_B = C_0$  at  $x = 0$ . The value of  $5.65 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$  was used as  $D_{AB}$  in the He/CO<sub>2</sub> system [32], and  $\tau$  was assumed to 1.54. This value was obtained at  $\varepsilon = 0.50$  using the following equation [33].

$$\tau = \sqrt{1 - \ln(\varepsilon^2)} \quad (6)$$

The velocity of the front line of the defluidized region ascertained from the pictures of the He/CO<sub>2</sub> system was

between 8 and 20 mm s<sup>-1</sup>. Although the other mechanism described in the He/Ar system and the gas-adsorption effect were not accounted for in the calculation, this result was nearly consistent with the velocity of the front line of the defluidized area observed in the He/CO<sub>2</sub> system shown in Fig. 9. This supports the mechanistic hypothesis that the defluidization was caused mainly by the adsorption of the second gas diffused into the emulsion phase from the channel.

### 3.4 Intensity of defluidization

We obtained the extent of defluidization area by dividing the number of pixels with red color by the number of pixels of the entire emulsion phase containing the defluidized region. The change in the extent of defluidization area with time after the gas switching is shown in Fig. 11. The extent and duration was affected by the combination of the first and second gases. In all cases, the defluidized region expanded just after the gas switching and reached a maximum value after 15–20 s. Subsequently, the fluidization quality was recovered gradually.

The defluidized region expanded more rapidly for H<sub>2</sub> than for He as the first gas. This was due to the small voidage in the emulsion phase when the bed was fluidized by H<sub>2</sub> gas as described above. In addition, the defluidization was more serious when CO<sub>2</sub> was used as the second gas. This was due to the gas adsorption on the porous particles and this effect was significant for the formation of defluidization. Fluidization was completely restored after 100 s even in the H<sub>2</sub>/CO<sub>2</sub> system in which the greatest influence was observed.

In previous studies [9–13], the intensity of the defluidization was expressed by the decrease in pressure drop across the bed. Fig. 12 shows the change in the pressure drop measured at the bed bottom in this study. Channeling occurred just after the gas switching and induced the decrease in pressure drop. The gas flow rate in the defluidized area would be negligible and a part of this area was supported by walls and a distributing layer. In all combinations, the pressure drop reached a minimum value after approximately 10 s. Subsequently, it increased and returned to the normal value gradually.

In spite of the opposite trend, the duration of the transient defluidization period was of the same order for each gas combination as shown in Figs. 11 and 12. The extent of defluidization area decreased from its peak at an approximately constant rate. On the other hand, the recovery rate of the pressure drop was relatively high. This means that the defluidized area was not supported only by walls, but this agglomerated area was supported by the fluidizing gas at the end of the recovery process.

## 4. Conclusions

The behavior of bubbles and bed voidage could be visualized by using a thin two-dimensional fluidized bed. Because the emulsion phase of the catalyst particles are fortunately expanded in fluidized beds with fine particles, the difference between the voidage of the fluidized region and defluidized region could be distinguished according to the transmittance of light. Based on this principle, the defluidization behavior occurred after the gas switching was successfully visualized using color gradation.

In the initial stage, the voidage of the emulsion phase was reduced and the defluidized region was formed at the bottom of the bed. The gas could not flow through the bed as bubbles, and channels were formed. The mechanism of the expansion of the defluidized region and the direction of the expansion were affected by the gas adsorption. When



the adsorptivity of the second gas was insignificant, the defluidized region expanded vertically after the initial formation of the defluidized region at the bottom of the bed. On the other hand, when the second gas was adsorptive, the defluidized region expanded horizontally as the diffusion of the second gas from channels to the emulsion phase.

As the surface of the defluidized region was eroded by gas-flow containing particles, the defluidized region was reduced and fluidization quality was restored. The method described in this paper will help in the analysis of the phenomena accompanying changes in bed voidage, such as the phenomenon after the gas switching in a fluidized bed.

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version.

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## Figure captions

**Fig. 1.** Relationship between gray-scale value and RGB values.

**Fig. 2.** Good fluidization quality after switching from Ar to He: (a) gray-scale picture, (b) colored picture.

**Fig. 3.** Defluidization phenomenon after gas switching from He to Ar: (a) gray-scale picture, (b) colored picture.

**Fig. 4.** Effects of the first gas on the emulsion phase voidage after 2 s of gas switching (a) from He to Ar and (b) from H<sub>2</sub> to Ar.

**Fig. 5.** Transient defluidization after gas switching from He to Ar. See also supplementary video 1.

**Fig. 6.** Change in the emulsion phase voidage at the bottom of the bed after gas switching from He to Ar.

**Fig. 7.** Two regions during the bed collapsing process.

**Fig. 8.** Effect of superficial gas velocity after gas switching on the duration of defluidization.

**Fig. 9.** Severe defluidization after gas switching from He to CO<sub>2</sub>. See also supplementary video 2.

**Fig. 10.** Calculation of CO<sub>2</sub> diffusion in a packed bed.

**Fig. 11.** Change in the extent of defluidization area after gas switching.

**Fig. 12.** Change in pressure drop through the bed after gas switching.