Chemical Engineering Science 96 (2013) 98–105

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Decrease in the fluidization quality of fluidized beds containing binary mixtures of different catalyst particles

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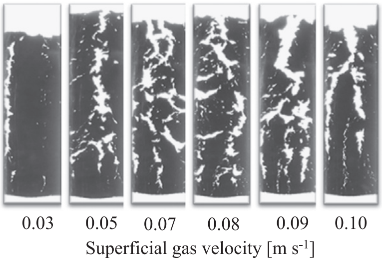
HIGHLIGHTS

- Fluidization quality drastically decreased when mixing catalyst and FCC particles.
- Gas channeling and irregularly-shaped voids were observed for mixed particles.
- The bed behavior was quite similar to that of Geldart's Group C particles.
- The collisions between the different types of particles caused the bipolar charging.
- The fluidization quality was reduced by the increase in cohesiveness.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 January 2013
Received in revised form
25 March 2013
Accepted 29 March 2013
Available online 10 April 2013

Keywords:
Fluidization
Fluidized bed
Chemical reactors
Agglomeration
Defluidization
Triboelectrification

ABSTRACT

Good fluidization was generally established when fine and light catalyst particles were fluidized. However, a large decrease in the fluidization quality was observed when two types of particles composed of different materials were mixed and fluidized, even though good fluidization was established with each type of particle alones. Formation of gas channeling was observed for binary mixtures of particles in a two-dimensional (2D) fluidized bed. In a three-dimensional (3D) bed, measurement of the pressure fluctuation showed that abnormal fluidization occurred due to mixing of the particles. In addition, measurements of the minimum fluidization velocity and emulsion phase expansion indicated that particle agglomeration occurred. The phenomena observed in the fluidized bed with the mixture of particles are quite similar to those observed for Group C particles. On the basis of the direction of the electrostatic charge, the particles were separated using electrodes installed above the fluidized bed surface. It was confirmed that one type of particle was positively charged and the other was negatively charged. Thus, it was concluded from these results that the bipolar charging caused by the difference in chemical composition of the particles induced the agglomeration phenomena and poor fluidization.

1. Introduction

Fluidized catalyst beds (FCBs) have been applied to many catalytic reactions (Kai et al., 1988; Ichihashi and Kitamura, 2002; Duvenhage and Shingles, 2002; Dente et al., 2003; Guerrero-Péreza et al., 2008; Go et al., 2010) and are still promising candidates as catalytic reactors for some new processes. However, it is necessary to establish good fluidization for the proper operation of catalytic reactors. For this reason, it is important to choose catalyst particles that possess, in addition to the properly high chemical activity, appropriate physical properties such as average size, size distribution, and particle density (Miyauchi et al., 1981). Many researchers have reported that fluidized beds of fine particles classified as Group A particles (Geldart, 1973) are suitable for catalytic reactors.

Miyauchi et al. (1981) investigated the particle properties used in industrial processes and showed that the physical properties of the particles used for successful commercial FCBs are practically limited to a narrow region. Particles with these properties are

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0009-2509/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ces.2013.03.057

categorized as Group AA (advantageous aeration) particles (Kai et al., 2004). For these particles, the optimum mean diameter is between 50 and 70 μm, the size distribution is wide, the fraction of large particles above 80 μm is 5–20 wt%, the fraction of small particles below 40 μm is 10–30 wt%, and the bulk density is typically falls between 400 and 1200 kg m^{-3}. This set of criteria has been a useful guide for many Japanese companies when designing catalysts. The mass transfer rate between the bubble and emulsion phases is increased by the high dispersion of small bubbles, and stable operation is ensured due to the small pressure fluctuations that occur when the particle properties are within the optimum range. In addition, the conversion and selectivity can be increased when the catalyst particles are in direct contact with the bubble phase gas and are suspended in the free board region (Tsutsui, 2004).

For industrial catalyst applications, it is possible to use a mixture of different types of catalyst particles. For example, the catalyst can be diluted by non-active particles, or multiple reactions can be controlled by mixing catalysts with different catalytic functions. In addition, particles possessing different surface conditions can coexist unintentionally in the system when catalysts are circulated between a reactor and a regenerator. When coke is formed during reactions over solid acid catalysts, the coke on the catalyst is burned in the regenerator. In the case of partial oxidation processes, metal oxide catalysts are reduced and change to the metal form in the reactor and then oxidized in the regenerator.

Kai et al. (2001) reported that the fluidization quality seriously decreased when two types of catalyst particles were mixed. These particles were both classified as Group AA and showed excellent fluidization quality when individually fluidized. Although it was assumed that electrostatic forces affected the fluidization quality, the details remain unclear. It is well known that the hydrodynamics of fluidized beds are strongly affected by triboelectrification (Mehrani et al., 2007a; Fang et al., 2008; Wang et al., 2009; Cheng et al., 2012; Sowinski et al., 2012). In particular, the symmetrical electrification resulting from mutual friction between the particles significantly affects the dynamics of the fluidized bed (Wolny and Kaźmierczak, 1993). However, very few studies on the fluidization of mixtures of multiple types of particles in FCBs have been published. Therefore, in this study, we observed the fluidization behavior of a binary mixture of particles and investigated the mechanism of the decrease in the fluidization quality. To evaluate the fluidization quality, the bubble behavior was observed in a 2D fluidized bed. In addition, the pressure fluctuations and the expansion of the emulsion phase were measured in a 3D fluidized bed.

2. Experimental

2.1. Particles

Two types of particles were used in this study: V_2O_3 catalysts supported on porous alumina particles (catalyst particles) and fluid catalytic cracking (FCC) catalysts (FCC particles). The FCC catalyst was a zeolite equilibrium catalyst. The physical properties of these particles are given in Table 1. Both the particles were Group AA particles and showed good fluidization quality when fluidized individually. The particles were dried at 400 K at least 12 h before each measurement. All experiments were performed at ambient temperature and pressure. Dried air was used as the fluidizing gas.

Table 1
Properties of the particles.

Particles	d_p [μm]	ρ_p [kg m^{-3}]	ρ_b [kg m^{-3}]	U_{mf} [mm s^{-1}]	U_{mb} [mm s^{-1}]
Catalyst	54.5	963	545	1.7	4.5
FCC	59.1	1484	851	3.5	6.7

2.2. Two-dimensional fluidized bed

A 2D fluidized bed with a cross section of 80 × 2 mm was used to observe the behavior of the bubbles and particles. The column was constructed from a transparent glass sheet having a thickness of 3.0 mm. Alumina balls (0.267 mm in diameter) were packed at the bottom of the column in order to distribute the fluidizing gas. Catalyst particles, FCC particles, and a binary mixture of the two (mass fraction of FCC particles was 0.5) were fluidized using dry air. The settled bed height was 0.18 m for all of the measurements. The fluidized bed was illuminated with back lighting and the motion of the bed was recorded using a video camera. The bed collapse behavior was also observed after the fluidizing gas was shut off when the bed was fluidized at $U_G = 0.1$ m s^{-1}.

2.3. Three-dimensional fluidized bed

A 3D fluidized bed was operated using a glass column with an inner diameter of 50 mm. Alumina balls (0.267 mm in diameter) were packed at the bottom of the column for gas distribution. The settled bed height was 0.3 m. In order to avoid the elutriation of the fine particles, a paper filter was installed at the top of the column. The fluidization quality was evaluated using the following measurements.

The minimum fluidization velocity, U_{mf}, and the minimum bubbling velocity, U_{mb}, were determined from the pressure drop and the change in the bed height when the gas velocity gradually increased and decreased. The measurements were performed just after the particles were loaded into the bed, and after the bed was fluidized for 5 min at $U_G = 0.025$ m s^{-1}.

Measurement of the pressure fluctuations was performed at 0.1 m above the surface of the gas distribution zone at $U_G = 0.02$–0.04 m s^{-1}. The analog signals from a pressure transducer were converted to digital signals using an A/D converter and then analyzed using a computer. The sampling time period and frequency of the pressure signals were 4 s and 1000 Hz, respectively. Therefore, the number of data points for each measurement, n, was 4001.

The average value of the pressure drop during the sampling period is described by the equation,

$$\Delta P_{av} = \frac{1}{n}\sum_{i=1}^{n} \Delta P_i \quad (1)$$

The intensity of the pressure fluctuations is defined by dividing the standard deviation of the measured pressure fluctuations (Shou and Leu, 2005; Chen et al., 2006) by the average pressure drop:

$$\delta_P = \frac{\sqrt{\frac{1}{n-1}\sum_{i=1}^{n} (\Delta P_i - \Delta P_{av})^2}}{\Delta P_{av}} \quad (2)$$

The autocorrelation coefficient, C_τ, was also obtained from the measured pressure data. This coefficient is a function of the time lag and is obtained as follows:

$$C_\tau(j) = \frac{\sum_{i=1}^{m} \Delta P(i)\Delta P(i+j)}{\sum_{i=1}^{m} \Delta P(i)^2} \quad (3)$$

Because the sampling frequency was 1000 Hz, the time lag was $j/1000$ s. In the analysis, the maximum time lag was 1 s. Therefore, j was changed from 0 to 1000, and the value of m was 3000.