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Fluidization of high-density particles: The influence of fines on reactor performance

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ABSTRACT

The effect of fines on the hydrodynamics of a gas–solid fluidized bed and ozone decomposition reaction was investigated using high-density iron-silicon (FeSi) particles with a particle density of 6690 kg/m^3. The addition of fines decreased the bubble size, dense phase voidage and reactor performance. The bubble size decrease is in accordance with reported literature; while, increase in dense phase voidage and reactor performance was found in the literature on less dense catalyst. The reactor performance was quantified using an apparent overall mass transfer parameter derived from fitting a two-phase model to the experimental data. The method allowed for reactor performance comparison despite the fluctuation in FeSi particle activity. Model fitting results suggest that smaller bubbles should improve mass transfer in addition to reactor performance. However, the decreased dense phase voidage with addition of fines counteracted the effects of smaller bubbles. Higher entrainment rate of the bed with fines was noted.

1. Introduction

Gas–solid fluidized beds are widely used in chemical and petrochemical industries owing to their efficient contact between the phases, resulting in good mixing and superior heat and mass transfer. Although the open literature contains numerous studies on fluidized bed hydrodynamics, very little is reported on highly dense particles [1,2]. The High-Temperature Fisher-Tropsch gas–solid fluidized bed is a unique example of an industrial process employing high-density particles with an estimated particle density ranging from 7000 to 9000 kg/m^3 [3,4].

The addition of fines in gas–solid fluidized beds has long been known to introduce desirable features into reactors. In industry, fines are mostly generated via attrition during normal reactor operation. For bubbling fluidized beds, a decrease in bubble size and an increase in dense phase voidage have been reported. This is in agreement with a longer collapse time for fluidized beds containing higher fines [5–8]. The mechanisms by which the addition of fines influences the hydrodynamics are still unclear. Some research suggests that in gas–solid fluidized beds the particles tend to form stable agglomerates when they are smaller than 20–40 μm [9]. From SEM images it was found that fine particles adhered to coarse particles or formed agglomerates [10]. Other effects of fines include the decrease of the velocity at which the turbulent fluidization regime starts [11–13]. Furthermore, the effect of fines on elutriation from fluidized beds was found to depend on the size and proportion of fines, as well as the gas velocity [9]. A more recent and systematic study has shown the influence of particle size distribution and the addition of fines on the hydrodynamic behaviour of Geldart A particles [14].

The change in hydrodynamics with addition of fines has also been proved to increase conversion in a catalytic reaction system. Yates and Newton [15] added 16% and 27% fines to a bed of Geldart A commercial oxidation catalyst, where fines were defined as particles <45 μm. The investigation found that reactor performance increased due to an increase in dense phase voidage. The voidage increase caused a shift in the gas flow pattern: more gas was flowing through the dense phase and less in the lean phase. Further experimental evidence was reported by Sun and Grace [16] on the disproportionate increase of fines contained in bubbles contributing to better chemical conversion with a wider particle size distribution [11]. The study was conducted with a narrow, bimodal and wide particle size distribution of Fluid Catalytic Cracking (FCC) catalyst. Fines were defined as particles smaller than 20% of the Sauter mean particle size.

The aim of this investigation was to observe the effect of fines on the reactor performance of dense particle fluidization. The velocity range included the bubbling and turbulent fluidization flow regimes, since the turbulent regime is generally associated with less interphase mass transfer restriction and, accordingly, improved reactor performance. Initial work on the technique of quantifying reactor performance was done using an FCC catalyst in a pseudo 2D column [17,18]. For this investigation the technique was refined and a different reactor modelling approach was used to gain insight into changes in reactor performance. High-density iron-silicon (FeSi) alloy particles were then used in a 3D cold bed. Conversion was determined using the ozone decomposition reaction, which is catalysed by naturally active FeSi particles. Additional hydrodynamic measurements included bubble size, solids concentration and entrainment rate.

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2. Experimental approach

2.1. Basic two-phase reactor model

As with most multiphase reactors, modelling of a fluidized bed is complex due to hydrodynamic behaviour. Two-phase theory is now generally accepted as the best modelling approach [19–21]. At its simplest, a fluidized bed reactor (FBR) can be described as a dense solids-rich phase with a lean solids-deprived phase bubbling through it. Reactant, mostly in the lean phase, is transported to the dense phase via mass transfer in which most of the reaction occurs. By assuming negligible gas flow through the dense phase and no solids content in the bubbling phase, the following mass balance can be done based on the solids volume of the catalyst:

$$C_i = u_b C_{i,B} + u_E C_{i,E} \tag{1}$$

$$A_{bed} u_b \frac{dC_{i,B}}{dV_s} = -K_O\left(C_{i,B} - C_{i,E}\right) \tag{2}$$

$$A_{bed} u_E \frac{dC_{i,E}}{dV_s} = -R_i(C_E) + K_O\left(C_{i,B} - C_{i,E}\right) \tag{3}$$

where R_i is the reaction rate. For first-order reactions it would be:

$$R_i(C) = k_r C \tag{4}$$

In this model the overall mass transfer coefficient (K_0) has the same units as k_r, which is based on the rate of total volumetric gas transfer per solids volume. It is important to understand that if K_0 is fitted to experimental data using this simple two phase model; it would be an apparent parameter; the concept is similar to an apparent reaction rate constant. When a reaction rate constant (k_R) is determined by fitting a Plug Flow Reactor (PFR) model to packed bed experimental data, flow effects or hydrodynamics would be incorporated in the value of k_R. The value of K_0 will be influenced by hydrodynamic behaviour which is not considered in the model. This apparent overall coefficient serves as an indicator of the reactor performance, irrespective of the catalytic activity of the bed. K_0 should not be seen as the actual mass transfer in the bed, but rather the measurement of reactor performance.

By taking into account the hydrodynamics of the reactor and the K_0 parameter, the generally used area-specific mass transfer coefficient can be calculated. K_0 should be multiplied by the ratio of solids to expanded bed and divided by the bubble fraction and the ratio of bubble volume to surface area [22]:

$$k_{be} = \frac{K_O\left(\frac{V_p}{V_{bed}}\right)}{\psi_B a_i} \tag{5}$$

where

$$\frac{V_p}{V_{bed}} = (1-\varepsilon_0) \tag{6}$$

$$\psi_B = \frac{u_o}{u_{br}} \tag{7}$$

For the bubble rise velocity (u_{br}):

$$u_{br} = 0.711\sqrt{gD_b} \tag{8}$$

From this discussion it is clear that k_{be}, is dependent on correlations, assumptions and model selection. A popular area-specific mass transfer correlation is that of Sit and Grace [23]:

$$k_{be} = \frac{1}{3}U_{mf} + \left(\frac{4D_m\varepsilon_{mf}U_{br}}{\pi D_b}\right)^{\frac{1}{2}} \tag{9}$$

2.2. Bubble measurement techniques

Two methods of bubble measurement were used in this investigation: an intrusive probe technique and a non-intrusive pressure measurement technique. The intrusive technique makes use of a voidage probe which detects a passing bubble due to a sudden drop in solids concentration around the probe. Using the probe-bubble contact time and a bubble rise velocity correlation, the bubble size is estimated. Karimipour and Pugsley [24] did a critical evaluation of all the available correlations, from which the most appropriate correlation was chosen for this system. The correlation of Werther (1978, included by the above authors) was used by taking the average between the Geldart A and B correlations; FeSi is at the boundary between A and B:

$$u_{br} = 0.934\sqrt{9.81D_b} \tag{10}$$

It is important to note that there is a difference between the average bubble size and the void length. To relate the void length distribution to an average bubble size, the equation of Liu and Clark [25] is required:

$$L_b = \left(\frac{2}{3}a(1+Q)^3 - aQ(1+Q)^2\right)D_b \tag{11}$$

where Q is the bubble wake shape factor.

With the probe-bubble contact time (t_1) and rise velocity correlation, a void length can now be determined:

$$L_b = t_1 u_{br} \tag{12}$$

and it can be shown that:

$$D_b = \frac{8.56}{\left(\frac{2}{3}\alpha(1+Q)\right)^3 - \alpha Q(1+Q)^2}t_1^2 \tag{13}$$

The non-intrusive technique of Beetstra et al. [14], which uses pressure measurement signals, was also used. The Power Spectral Density (PSD) function of the signals is used to decompose pressure fluctuations into global bed phenomena and phenomena in the vicinity of the pressure probe. These local phenomena are caused by the passing bubbles/voids. Two pressure probes are required: one in the plenum chamber or directly above the distributor, and a second at a height in the bed where bubble measurement is desired. The PSDs of both pressure probe signals are compared and the incoherence of the two signals relative to each other is calculated. The standard deviation of this incoherence (σ_i) is a measure of the average bubble/void size. This is directly proportional to the bubble size in the following manner:

$$D_b \propto \frac{\sigma_i}{\rho_b g} \tag{14}$$

which means:

$$D_b \propto \frac{\sigma_i}{\rho_p(1-\varepsilon_0)g} \tag{15}$$

2.3. Equipment and method

The investigation was conducted using a 0.14 m inside diameter acrylic column of height 5.5 m. To return entrained solids to the bed, a system with two external cyclone returns was used. Filter bags were installed after the cyclones and weighed before and after experiments. A triangular pitch perforated plate distributor with thirty 2 mm holes was used with an open area of 0.61%. To prevent solids weepage, a porous cloth was placed below the distributor. The particles used for these experiments were a high-density FeSi