Computational study of the bubbling-to-slugging transition in a laboratory-scale fluidized bed

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Abstract

We report results from a computational study of the transition from bubbling to slugging in a laboratory-scale fluidized-bed reactor with Geldart Group B glass particles. For simulating the three-dimensional fluidized-bed hydrodynamics, we employ MFIX, a widely studied multi-phase flow simulation tool, that uses a two-fluid Eulerian-Eulerian approximation of the particle and gas dynamics over a range of gas flows. We also utilize a previously published algorithm to generate bubble statistics that can be correlated with pressure fluctuations to reveal previously unreported details about the stages through which the hydrodynamics progress during the bubbling-to-slugging transition. We expect this new information will lead to improved approaches for on-line

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reactor diagnostics, as well as new approaches for validating the results of computational fluidized-bed simulations with experimental measurements.

1. Introduction and Background

Gas-solid fluidized-bed reactors are widely used in the chemical industry, including biomass conversion [1-4], petroleum refining [5], and pharmaceutical [6, 7] and commodity chemicals production [8]. For this reason, there is widespread interest in establishing a comprehensive understanding of the gas-solid hydrodynamics to optimize processes in which fluidized-bed reactors are key components. Three of the most important hydrodynamic states or flow regimes in fluidized-bed reactors are referred to as freely bubbling, slugging, and turbulent fluidization. Each regime has distinct physical characteristics that produce widely different levels of heat and mass transfer between the gas and solid phases [3, 9]. Thus it is important to distinguish how the fluidized-bed design and operating conditions correlate with fluidization state. Where possible, identifying on-line process measurements monitoring the fluidization state can be especially useful. This study focuses on understanding how to use pressure measurements to distinguish between free bubbling and slugging.

The free-bubbling state is typically the first condition encountered as the gas flow is increased above the minimum fluidization state [10] in beds of solids within the Group B category of the Geldart's classification scheme [11]. In this regime, pockets of gas (bubbles) form near the bottom of the bed and rise upward until they reach the surface, at which point they erupt. The appearance of the fluidized bed in this case is typically described as similar to that of a boiling liquid. The solids and gas motion associated with the rising bubbles [12] leads to enhanced heat and mass transfer, which is one of the primary advantages for carrying out reactions between fluids

and solids in bubbling fluidized beds. Many descriptions of this flow condition are available in literature with comparisons between liquid-gas and solids-gas systems [13-15].

As the gas bubbles rise in a fluidized bed, they typically grow larger due to coalescence and decreased pressure. When the gas flow is sufficiently high and/or the bed sufficiently deep, slugging occurs. Slugging has long been recognized as an area of concern because it is characterized by the formation of large gas bubbles that produce intense oscillations of the bed solids. Criteria have been proposed for the conditions necessary for slugging to develop, such as the following: (1) the static bed height must exceed a critical height; (2) the superficial gas velocity must exceed a minimum (slugging) velocity; and (3) bubbles must be able to grow to a size that approaches the diameter of the fluidized-bed vessel [16, 17]. Typically, many observers report that slugging evolves from the freely bubbling fluidization state as superficial gas velocity is increased, but it is often unclear in the literature just where (or if) there is a clear point of demarcation between free bubbling and slugging. Minimum slugging correlations [18-20] developed from experiments also result in very different values for inlet velocities. Furthermore, some studies [21] have even mistaken the peak in standard deviation as the slugging transition.

Early investigators [18-20, 22-25] developed minimum slugging correlations based on experimental observations. Although these relations are useful for understanding general trends, they relied heavily on subjective visual observations and qualitative features. More recent studies [26-35] have utilized direct quantitative measurements of pressure and void fraction to monitor the spatiotemporal behavior of bubbles. Fan et al. [27], for example, reported a decrease in bubble rise velocity associated with slugging based on cross correlation of pressure measurements in the upper portion of the bed. Similarly, Lee et al. [30] and Saxena and Rao [36] investigated the slugging transition by analyzing pressure measurements from the upper half of a laboratory fluidized bed.

[37-41], on the other hand, used electrical capacitance tomography (ECT) to image changes in bubbles associated with the slugging transition. In all these experiments, however, the complex nature of the free bubbling to slugging transition has not been found to be amenable to a clear and consistent physical model.

Computational fluid dynamics (CFD) has a long history of application for dynamic simulation of fluidized beds. Ideally, it should be possible to incorporate sufficient details of the physics of granular and multi-phase flows to replicate all of the salient hydrodynamic features of slugging fluidized bed reactors. The process of capturing all of the critical aspects of the physics at multiple scales remains extremely challenging however, and there are many opportunities for additional improvements. Examples of the challenges and complexities involved in CFD simulations of slugging fluidized beds are highlighted in recently published work such as that by Ichiki et al. [42], Pain et al. [43, 44], Zhang and Yu [45], Reuge et al. [12, 46], Goldschmidt et al. [47], Loha et al. [48, 49], Fede et al. [50], Xie et al. [51], Li et al. [52], and Bakshi et al. [53, 54]. We emphasize here that our goal is not to attempt to develop and improved any specific model of fluidized bed physics but rather to pursue a better understanding of the basic quantitative trends underlying bubble dynamic behavior as it is predicted by such simulations. We specifically target the dynamics of bubbles and the onset of slugging, because these are known to be critical features that are central in determining the performance of practical bubbling bed reactors. Ultimately, we also are concerned with identifying how bubble patterns can be used to experimentally test the validity of the computational simulations and as a practical monitoring tool (i.e., which bubble measurements should be most useful for validation and on-line diagnostics).

The primary objective of this study is to improve understanding of the transition between free bubbling and slugging by probing the physical details of the process revealed in computational

simulations of a laboratory-scale bubbling fluidized bed. We hypothesize that computational studies of this type, as long as they are done with proper care, can provide information that is not directly available from experimental measurements. With this additional information, it should be possible to develop methods based on high speed pressure measurements to detect the approach of slugging in both laboratory and industrial reactors before it becomes problematic. This information can be useful in improving the physical models on which computational simulations are based.

2. Technical Approach

2.1. Fluidized bed simulation conditions

To reflect a lab-scale reactor of current relevance, we assumed the geometry of an experimental laboratory reactor used for biomass processing research at the National Renewable Energy Laboratory (NREL). A schematic of the reactor is shown in Fig. 1. The inner diameter D_r and height of the reactor H_r are 0.0508 and 1.27 m, respectively. However, for simulation purposes, the computational domain was reduced to a height of 0.4 m. Operating conditions were chosen to match baseline experiments at NREL and are listed in Table 1. Initially, glass particles with diameter d_s of 2.5×10^{-4} m and density ρ_s of 2484 kg/m^3 were set at a static bed height of $H_o = 0.2032$ m, with an initial void fraction of 0.4. The glass particle-particle properties were defined with a coefficient of restitution of 0.8 [12] and angle of repose at 30° . The particle-wall collision specular coefficient was set to 0.6, however when normalized slip velocity goes to zero, it is calculated internally using a relation developed by Li et al. [52, 55]. The reactor outlet is open to pressure at 255 kPa. Each simulation was initiated by uniformly adding nitrogen gas through the reactor bottom with a mass flow inlet based on superficial gas velocity U , as a multiple of the minimum fluidization gas velocity U_{mf} . The minimum fluidization velocity was calculated at 0.03

m/s, and the Syamlal-O'Brien drag-model [56] parameters (see Section 2.2) were assigned based on the calculated U_{mf} and operating conditions. U_{mf} was calculated using the Richardson equation [10] and values in Table 1 together with nitrogen density using the ideal gas form and viscosity using the NASA polynomial for transport properties.

Simulations were conducted with all operating parameters held constant and only inlet velocity, U , was varied from 1.25 to $2.75U_{mf}$ to identify the onset of the bubbling-to-slugging transition and fully developed slugging. The shown herein are for a single static bed depth, and the transition flow and location should be expected to change with bed height, diameter, and particle characteristics. However, the general utility of this methodology should be expected to hold.

Table 1. Operating conditions

Definition	Units	Experiment
Particle diameter	m	2.5×10^{-4}
Particle density	kg/m^3	2484
Bulk density	kg/m^3	1552
Temperature	K	773
Pressure	kPa	289
Fluidizing N_2 (range)	kg/s	$8.46 - 18.6 \times 10^{-5}$

Fig. 1. Schematic diagram of the simulated bubbling bed

2.2. Computational Methods

We employed computational fluid dynamics (CFD) [57] to simulate the hydrodynamics in the bubbling bed reactor described above over a range of fluidization velocities. Numerous fluidized-bed researchers have found CFD to be a useful complement to experimental measurements [52, 53, 58, 59]. One of the benefits of CFD is that it can provide spatiotemporal details about pressures, velocities, and concentrations that are either impossible or extremely difficult to obtain experimentally. CFD has also been employed in numerous studies of gas-solid fluidized beds [58, 60], but very limited number of CFD studies have addressed the bubbling-to-slugging transition [17, 61, 62].

The specific CFD implementation used in this study employed the Eulerian-Eulerian computational Two-Fluid Model (TFM) [63, 64], which approximates the flowing phases as interpenetrating continua. This is in contrast to numerical simulations that resolve discrete particles

or molecules [62, 65-67]. While the TFM approach does not resolve individual particles, it has been demonstrated to reproduce major hydrodynamic features, including bubbles [12, 51, 53]. Detailed information on the TFM formulation can be found in Gidaspow [63].

To carry out our simulations, we utilized MFiX (Multiphase Flow with Interphase eXchanges), which is an open-source CFD software developed primarily at the National Energy Technology Laboratory [68]. The gas phase was simulated as incompressible, and stress tensors for the gas and solid phases were related to shear stress using Newton's law. To model solids transport properties, such as solids pressure and viscosity, the kinetic theory of granular flow [68] together with the Schaeffer frictional stress tensor formulation [69] and the sigmoidal blending stress function [51, 60, 70] were employed to relate the computed solids temperature with solids transport properties. Furthermore, the gas-solid momentum transfer used the Syamlal-O'Brien correlation [56] for the drag model. The discretization scheme utilized a finite-volume approach with a staggered 3D grid [71]. Scalar values, pressure and void fraction, were stored in the cell center, while velocities were computed on the cell surfaces. Additionally, second-order discretization was utilized using the superbee approach which improved convergence and accuracy of the simulation. A modified SIMPLE approach [71] is also used and improves speed and stability through variable time stepping, solid volume fraction correction, and solids-pressure evaluation. The no-slip condition was applied to the gas phase on the side walls, while the Jackson and Johnson partial-slip wall boundary condition [72] was applied to the solid phase.

To characterize the spatiotemporal dynamics generated by MFiX, we tracked detailed variations in void fraction and pressure as time series at each computational grid point (no reactions or heat or mass transport were included). As explained below, these raw time series were then further processed to produce simulated (virtual) measurements of the bubble patterns and local

pressure fluctuations. We then analyzed and compared these time series at each axial location in the bed to understand their correlation with the bubbling and slugging states.

A general concern for multiphase flow CFD simulations is establishing a computational grid size that is sufficiently refined so spatiotemporal dynamics no longer depend on grid resolution (i.e., grid independence). We initially used a 2D fluidized bed simulation to study the impact of the mesh cell size in the x and y-directions on the statistical convergence of the simulated time series. Fig. 2 illustrates the effect of mesh cell aspect ratio (AR), $AR = \Delta y / \Delta x$, on the simulated pressure time series, standard deviation, skewness, and kurtosis. These results show mean pressure profiles are not sensitive to cell aspect ratio. However, the higher-order moments are sensitive to cell aspect ratio, which capture higher dimensional dynamics associated with bubbles and mixing. Details about the cell geometries and run times are provided in [Supplementary Table S1](#). The 40-second 2D simulation with $AR=1$ at $5U_{mf}$ was further refined to show grid independence ([Supplementary Fig. S1](#)) from 15×120 cells to 30×240 cells which resulted in run times of 16.5 and 122 hours, respectively.

To account for higher dimensional dynamics [51], a 3D fluidized bed was simulated at $2.75U_{mf}$ with a fine cylindrical mesh where $\Delta r = \Delta y$, resulting in a uniform mesh with 15 cells in the radial direction and 240 cells in the axial direction, and with 6 azimuthal cells. Skewness and kurtosis did not change as the number of cells in the azimuthal direction was changed to 6, 12, and 24 ([Supplementary Fig. S2](#)). Based on these results, we selected 6 cells in the azimuthal direction for continued simulations.

Fig. 2. Pressure statistics convergence at different grid aspect ratios, $AR = \Delta y / \Delta x$.

2.3. Statistical Methods

To quantify spatiotemporal dynamic transitions, we determined the statistical properties of pressure and void fraction time series at different axial positions in the bed. We specified normalized axial locations as H/H_o by specifying 10 equally spaced locations H_i up to the static bed height, H_o . We typically examined the bubble statistics at the horizontal planes at 0.1 up to 1.0 H/H_o . We then generated time series by interpolating the pressure and void fraction measurements from the nearest grid points at each simulation time step (0.01 s). For pressure, all the grid point values in a plane were then averaged together to create a single average pressure time series for each bed axial location. Experimental groups [73, 74] have found pressure measurements are influenced by a volumetric region close to the pressure port, and the analysis method used here captures dynamics in a defined volumetric region composed of cells near the probe measurement point.

To assist in understanding the physics involved, we also processed the void fraction time series at each grid point with MS3DATA, a special algorithm recently developed by Bakshi et al

[53], to resolve bubble sizes and numbers in fluidized beds. MS3DATA is a code developed in MATLAB and identifies bubbles in a five-step process [54]: data collection, bubble detection, conflict resolution, bubble properties and Lagrangian velocimetry. Initially the code collects void fraction data and bubble boundary resolution. It is followed by applying the user specified void-fraction threshold criteria and bubble linking, to define regions of contiguous subthreshold void fraction. The code then performs bubble conflict resolution by assigning unique bubble numbers to every grid cell. Bubble properties such as location, size, span, and shape are then assigned. The code can also track bubbles across successive time frames, but this analysis was not performed in this investigation.

For consistency, only cells with void fraction > 0.7 [39, 40, 53, 75] were considered as representing bubbles, and we found results did not change significantly by selecting a slightly different cut-off value (Supplementary Fig. S3). Bubble-counting domains were defined as axial locations up to the halfway points of adjacent axial planes (e.g, the volume between 0.85 and 0.95 H/H_o for counting at 0.9 H/H_o). Bubbles whose centroids resided within each axial domain were used by MS3DATA to calculate bubble size and number. Within each axial domain, bubble diameter d_b was evaluated using the linked bubble volume V_b according to $d_b = \sqrt[3]{6V_b/\pi}$. More details of the bubble-statistics algorithm in MS3DATA can be found in Bakshi et al. [76]. Initial tests showed pressure and bubble statistics became statistically stationary after 7 seconds of physical time (Supplementary Fig. S4 and S5). Therefore, although the simulations were sampled for 40 seconds at 100 Hz, the first 10 seconds were discarded, resulting in time series ranging from 10 to 40 sec for pressure and bubble statistics.

Numerous statistical descriptions, some of which are based on concepts from nonlinear and complex systems theory, have been proposed for characterizing time-series measurements from

fluidized beds [15, 26, 30, 73, 77-83]. For this study we elected to concentrate on more traditional statistical measures to simplify the analysis and development of relationships between the pressure and bubble patterns. These statistics included histograms of the time-series values as well as quantitative characterizations utilizing the mean, standard deviation, skewness, and kurtosis (Supplementary Eq. (S.1)–(S.4)). We also utilized the autocorrelation and cross-correlation (Supplementary Eq. (S.5) and (S.6)) functions [84] to evaluate relationships between different measurements at different points in time.

In some cases, the time series were normalized by their mean to highlight certain features. These cases are described in the Results and Discussion section. We also utilized the method (Supplementary Eq. (S.7)) proposed by Scott [85] to select the best bin sizes for constructing frequency histograms.

3. Results and Discussion

The visual appearance of the bubbles generated by MFiX was observed graphically in terms of void fraction iso-surfaces using Paraview [86] as illustrated in the following section below. To systematically quantify the simulated bubble patterns, we evaluated bubble statistics derived from the MFiX void fraction output using MS3DATA. We then evaluated the pressure time series features at each level to determine how they relate to the observed bubble behavior.

3.1.1. Bubble characteristics at low gas flows (free bubbling)

Fig. 3. (a) Time-average vertical profiles of the simulated bubble size and bubble count (concentration) per frame under low gas flow conditions ($U=1.25U_{mf}$); (b) instantaneous snapshot of bubble iso-surfaces; (c-e) bubble size histograms at $H/H_o = 0.3, 0.6$, and 0.9 , respectively.

Fig. 3(a-e) illustrate typical bubble characteristics we observed in low gas flow simulations. Fig. 3(a) shows the predicted time-average profiles for bubble size and concentration at a gas flow corresponding to $U = 1.25U_{mf}$ as determined from the MFiX output with MS3DATA [76]. Fig. 3(b) illustrates an example instantaneous snapshot of bubbles predicted for this same condition from the MFiX output with Paraview. Fig. 3(c-e) reveals characteristic bubble size growth as the bubbles move from lower to higher levels. This is consistent with the expected effects of bubble-to-bubble coalescence and reduced pressure [87, 88]. However, MS3DATA results indicate the

number of bubbles increase with height until reaching the splash (eruption) zone [88]. We conjecture that this growth in numbers might be explained as the result of bubble splitting and/or the increase in gas flow above U_{mf} due to the reduced pressure. Bakshi et al. [54] published work which utilized MS3DATA on larger-diameter fluidized beds (15, 30, 50 and 70 cm). In these studies, they observed a larger number of bubbles (bubble count/frame) in the bottom relative to the top of the bed. This was also the trend we saw for reasonable assumptions about the void fraction cutoff limit at high gas flows as indicated in [Supplementary Fig. S3](#). However, we emphasize that direct comparisons between the reactors simulated by Bakshi et al. and the reactor in this study are likely to be misleading because of the different reactor sizes and particle properties. In particular, the larger diameters of the Bakshi et al. reactors (making it possible to generate larger bubbles with reduced wall drag) and their larger particles (putting them on the Geldart B/D boundary) would be expected to significantly influence the bubble behaviors.

Although the simulated bubbles described above appear to reflect the expected trend in bubble size, it is not clear to us that the predicted bubble shapes are consistent with direct visual observations reported by experimental studies. In particular, it appears that the simulated bubble shapes are less regular and somewhat flattened compared to images from experimental ECT and x-ray observations [89-91] at similar fluidization states. Additionally, we did not expect the number of bubbles to increase with height as much as indicated by the MS3DATA results. Instead, this particular trend may provide a good way to test the soundness of bubbling bed simulations. If experiments can confirm that the predicted bubble number trends are indeed wrong for these conditions, it may reveal an inherent shortcoming in the two-fluid approximation that needs to be resolved in future studies. Based on other investigations [12, 48, 49, 53, 92] it might be possible to address this shortcoming by tuning one or more MFIX parameters based on experimental

measurements [12, 31, 35, 47, 54]. On the other hand, it may be that the two-fluid simulations are revealing a feature of bubbling beds that has not been previously recognized.

3.1.2. Bubble characteristics at high gas flows (fully developed slugging)

At much higher gas flows (e.g., $U = 2.75U_{mf}$) the simulated time-average bubble profiles change dramatically as depicted in Fig. 4(a), where the average bubble concentration reaches a maximum much lower in the bed and then drops precipitously with height. On the other hand, the average bubble size near the top of the bed grows until it approaches 60% of the bed diameter. At $H/H_o \sim 0.5$ the bubble size and bubble concentration curves exhibit significant changes in slope, which we conjecture may be the result of bubble coalescence events similar to those observed in experiments by [35, 88]. It is important to note that mean bubble number per frame reflects the average volumetric bubble number concentration in an axial plane. So it is also useful to consider supplementary Fig. S3, which depicts trends in the bubble count/frame vs. axial location. When viewed this way, our results show that at high gas flows, the region of the bed below $0.4 H/H_o$ exhibited a decreased bubble count per frame as the void fraction cutoff value was decreased from 0.7 to 0.55. We also note, however, that in the lower region of the bed, the simulated bubbles at high gas flow had less distinct boundaries, making it more difficult to discriminate between separate bubbles (and thus accurately count bubbles) when the cutoff void fraction used to recognize bubbles is decreased to 0.55 or below. As long as cutoff values of 0.7 and 0.6 were specified, the general axial trend in bubble frequency did not change much.

In Fig. 4(b) we see that the largest simulated bubbles in the upper part of the bed begin to assume ogive (bullet-like) shapes, which nearly fill the bed cross section, fully developed slugging bubbles. We conjecture that as these bubbles grow larger, the increased wall drag should slow their

rise velocity, allowing smaller trailing bubbles to catch up and coalesce with them, making them larger still. This suggests that the bubbling-to-slugging transition is inherently a type of avalanching or critical transition process [26].

Fig. 4. (a) Time-average vertical profiles of the simulated bubble size and bubble count (concentration) per frame under high gas flow ($U = 2.75U_{mf}$); (b) instantaneous snapshot of bubble iso-surfaces; (c-e) bubble size histograms at $H/H_o = 0.3$ (c), 0.6 (d), and 0.9 (e).

Bubble pattern differences are also evident in the time-average histograms of bubble size. Near the bottom of the bed (Fig. 4(c)) the bubble-size histogram has a positive tail that grows more pronounced with height and transitions to a bimodal shape near the top of the bed (Fig. 4(d) and (e)). We conjecture that this bimodality results from repeated coalescence events between some but not all of the rising bubbles, reflecting a global bifurcation process consistent with the observations of visualization experiments [22, 39, 40]. Similar bifurcation phenomena have been observed in bubble columns of highly viscous fluids during bubble coalescence [82, 93], implying

that this might be an example of a more general bubble phenomenon [94]. Although liquid-gas and solid-gas bubbles are governed by different physics [13], it seems plausible that there could be similarities in their regime transitions [13-15].

3.1.3. Bubble patterns at intermediate gas flows (between free bubbling and fully developed slugging)

Fig. 5. Bubble shape evolution at 0.03 s time steps at intermediate gas flow ($U = 1.75U_{mf}$). The highlighted regions compare two different bubble size clusters at different locations.

For gas flows between the high and low limits, the simulated bubbles exhibited intermediate emerging characteristics as depicted in the example time sequence in Fig. 5. Under these conditions, we observed repeated instances of smaller bubbles merging together to form larger

bubbles. While these larger bubbles initially accelerated relative to the small bubbles (note the larger travel distance of bubble cluster A compared with bubble cluster B), the acceleration became retarded when the bubbles grew sufficiently that their edges approached the wall. These interactive alterations of bubble size and speed appeared to be the essential processes behind the emergence of slugging in the MFiX simulations.

Fig. 6((a) and (b)) illustrate the trends in the vertical time-average bubble size and concentration profiles extracted with MS3DATA[76] from the MFiX output over the entire flow range between 1.25 and $2.50U_{mf}$. From these, it can be seen that the average bubble diameters are always largest near the top of the bed for all gas flows, although the difference in bubble size between the top and bottom increases with increasing flow. The biggest increase in this difference appears to happen for flows just above $1.75U_{mf}$. On the other hand, the trend in bubble concentration (Fig. 6(b)) appears to undergo an even more distinct transition when the gas flow exceeds $1.5U_{mf}$. Above $1.5U_{mf}$ the bubble concentration in the lower bed begins to exceed that in the upper bed. The upper bed also reaches a maximum bubble concentration. Careful observations of the detailed bubble sequences ([Supplementary video 1](#)) generated by MFiX indicate that this is the gas flow condition in which the intermediate emerging bubble characteristics from Fig. 5 begin to appear near the top of the bed. Thus, this is the beginning of the bubbling-to-slugging transition. Similarly, at $2U_{mf}$, bubble concentration first reaches a minimum near the top of the bed, indicating large, ogive bubbles first begin to emerge near the top of the bed. Bubble concentration at higher gas flows appears to converge to this same minimum bubble concentration near the top of the bed, indicating the end of the bubbling-to-slugging transition and the beginning of the fully developed slugging regime.

Fig. 6. Predicted time-average bubble diameter (a) and bubble count (concentration) per frame (b) profiles in the bed for a range of gas flows.

Additional perspectives on the intermediate-flow bubble patterns are revealed in the time-average bubble-size histograms in Fig. 7. In this figure, two demarcation lines are drawn: the first (marked in orange) indicates the approximate vertical locations and gas flows where significant bimodal features and ogive bubbles begin to appear, and the second (marked in green) indicates where bimodality and ogive bubble shapes become dominant features. We suggest that these lines outline a region of gas flow and bed height, within which, the free bubbling to slugging transition occurs. Bubble size histograms (Fig. 7) and bubble concentration (Fig. 6(b)) illustrate the bubbling-to-slugging transition occurs between 1.5 to $2U_{mf}$, also corresponding to the bubble histograms at the top of the bed.

Fig. 7. Bubble-size histograms at different H/H_o and U/U_{mf} .

3.2. Trends in predicted pressure fluctuations

As might be expected, the two-fluid MFiX simulations predict a complex but significant relationship between the bubbling transition patterns and pressure variations. As described above, the observed bubble behavior varied significantly with axial position, and this was mirrored by corresponding changes in the relationship with pressure. Some major features of this relationship and how it varied with position are summarized below.

3.2.1. Pressure fluctuations at low gas flow (freely bubbling)

Fig. 8(a-h) depict key characteristics of the pressure-fluctuation profiles predicted by the simulation at low gas flow (free bubbling at $U = 1.25U_{mf}$). As expected, the mean pressure values

at each level dropped monotonically in moving from the bottom to the top of the bed (Fig. 8(a)). Consistent with the previously described bubble growth trends with height, the standard deviation of the pressure fluctuations also grew with height (Fig. 8(a)). Similarly, skewness (S_p) and kurtosis (K_p) variations with height (Fig. 8(b)) were also consistent with changes in the pressure-fluctuation distributions with height (Fig. 8(f-h)) and reflect major shifts in the bubble-size distributions. We conjecture that these changes in distribution are associated with changes in the bubble shape and coalescences described above and expect that these statistics should contain useful diagnostic information [73].

Fig. 8. Pressure fluctuation patterns at $1.25U_{mf}$: (a) time-average pressure ($\bar{P}$) and standard deviation (σ_p); (b) skewness (S_p) and kurtosis (K_p); normalized pressure time series and pressure probability distributions at $H/H_o = 0.3$ (c,f), 0.6 (d,g), 0.9 (e,h).

3.2.2. Pressure fluctuations at high gas flow (fully developed slugging)

At high gas flow ($U = 2.75U_{mf}$) the predicted pressure-fluctuation time series and statistical profiles exhibit significant changes as illustrated in Fig. 9(a-h).

Fig. 9. Pressure fluctuation patterns at $2.75U_{mf}$: (a) time-average pressure ($\bar{P}$) and standard deviation (σ_p); (b) skewness (S_p) and kurtosis (K_p); normalized pressure time series and pressure probability distributions at $H/H_o = 0.3$ (c,f), 0.6 (d,g), 0.9 (e,h).

As in the low-flow condition, sharp positive and negative spikes were visible in the pressure time series, resulting in large changes in standard deviation, skewness, and kurtosis values related to the histograms with height. We speculate that these spikes reflect the influence of local, low-amplitude wake and drift bubble passage effects as suggested by Rudisuli [88], as well as bubble coalescence events and the downward propagation of global pressure waves originating from bubble eruption, bed expansion, and contraction [88]. We also suspect that the bimodality in the pressure fluctuations near the top of the bed may be correlated with the bimodal bubble-size distributions described above. Other investigators [73] have proposed that these changes in the skewness and kurtosis of pressure fluctuations might be used as a bubble diagnostic [95] that is

not as easily observable [96] in the mean and standard deviation. Also, the standard deviation of pressure decreases at higher H/H_o due to the lower hydrostatic pressure and bubble-eruption events at the bed surface, which equalize pressure with the freeboard.

3.2.3. Pressure fluctuations at intermediate gas flows (between free bubbling and fully developed slugging)

As with the bubble patterns, pressure fluctuations at intermediate gas flows exhibited characteristics between those observed at the low and high flow limits in this study. An example of these intermediate characteristics is depicted in Fig. 10(a-h).

Fig. 10. Pressure fluctuation patterns at $1.5U_{mf}$: (a) time-average pressure ($\bar{p}$) and standard deviation (σ_p); (b) skewness (S_p) and kurtosis (K_p); normalized pressure time series and pressure probability distributions at $H/H_o = 0.3$ (c,f), 0.6 (d,g), and 0.9 (e,h).

The significant changes occurring in the pressure fluctuations between the low and high gas flow, indicate that pressure histogram statistics might serve as useful diagnostics for the freely

bubbling to slugging transition as illustrated in Fig. 11 for skewness and Fig. 12 for kurtosis. This appears to be especially true for measurements from the upper region of the bed.

Fig. 11. Variations of skewness (S_p) in the predicted pressure fluctuations with respect to gas flow at different bed heights (a); S_p with respect to U at $H/H_o < 0.7$ (b); and S_p with respect to U at $0.7 < H/H_o < 1.0$ (c).

The relative importance of the skewness and kurtosis changes in these simulated pressure fluctuations appears to be consistent with the experimental observations of Lee et al. [30], who noted that a minimum in skewness in the upper bed ($0.65 < H/H_o < 1.0$ [97]) appeared to correlate with the bubbling-to-slugging transition. Based on our simulation results, the peak of S_p at the lower part of the bed ($H/H_o < 0.7$) might also be a characteristic indicator of the onset of the transition to slugging. Lee et al. [30] also found that an increase in K_p for pressure fluctuations in the upper section of the bed correlated with the bubbling-to-slugging transition. This appeared to be consistent with our simulations, as shown in Fig. 12(a).

Fig. 12. Variations of kurtosis (K_p) in the predicted pressure fluctuations with respect to gas flow at different bed heights (a); K_p with respect to bed height at various gas flows (b).

Data from Fig. 12(a) were re-plotted (Fig. 12(b)) to illustrate trends in K_p which relate to the fully developed slugging bubbles observed in section 3.1.3. At $2U_{mf}$ near the top of the bed, K_p first becomes negative, which appears to indicate the beginning of fully developed slugging. As gas flow increased, the K_p curves shifted to lower locations in the bed, as similarly predicted by the bubble statistics. Furthermore, to supplement this observation, [Supplementary Fig. S6](#) first illustrates an inflection point in standard deviation at $2U_{mf}$. This observation is consistent with experimental observations by Daw et al. [26] where the “maximum stable slugging” conditions occurred where there was an inflection point in the standard deviation curve.

3.2.4. Time scale information in the pressure fluctuations

Besides exhibiting changes in statistical distribution with gas flow, the simulated pressure fluctuations also exhibited changes in time scale that correspond to shifts in bubble behavior. This is illustrated by changes in the pressure time series autocorrelation function, as depicted in Fig. 13. As the simulated gas flow increased, the autocorrelation became more periodic (especially at higher levels in the bed), revealing the impact of a few, large bubbles. The time scales of the largest

oscillations at high flow were typically 0.3–0.5 s, corresponding to the frequency range associated with the large ogive bubbles in the upper bed. This implies that time-scale variations, such as observed in nonlinear-dynamics [15, 26, 28, 79, 83], as well as statistical distribution variations in the pressure fluctuations, such as those presented here, should be useful diagnostic tools for monitoring the bubbling-to-slugging transition. As noted above however, interpretation of these diagnostics will depend greatly on where (H/H_o) the pressure measurements are made.

Fig. 13. Example autocorrelation functions for the simulated pressure fluctuations at 3 vertical locations for low, intermediate, and high gas flows.

3.3. Cross-correlations between bubble patterns and pressure variations

Cross-correlation provides another method for quantitatively confirming the predicted physical connection between the void fraction (bubble behavior) and pressure fluctuations. Example results for the two-fluid MFiX simulations are depicted in Fig. 14, where the cross-correlation of void fraction with respect to pressure from bubbling to fully developed slugging at 0.9 is compared with the void fraction and pressure differential time series. As can be seen, the cross-correlation between these time series begins rising rapidly with gas flow and always reaches

a maximum just below the static bed height. In the future, we expect that cross-correlations such as this can provide a useful way to compare computational simulations with experimental measurements in order to validate and/or refine computational fluidization models.

Fig. 14. Maximum absolute magnitude of the cross-correlation between pressure and void fraction time series with respect to H at different U .

3.4. Comparison of the predicted trends with previous correlations

As discussed earlier, an important motivation for studies of this type is the apparent inconsistency of slugging predictions from correlations in literature. Fig. 15 below illustrates this for the present case by comparing the predictions of slugging correlations proposed by Stewart and Davidson [20], Baeyens and Geldart [18], Broadhurst and Becker [19] and Shaul et al. [98] (Supplementary Eq. (S.8)–(S.10)) with the gas flows predicted by MFiX for the initial onset of the bubbling-to-slugging transition and complete transition to slugging, fully developed slugging. As can be seen there is considerable variation among the predictions from these correlations about the gas flow at which slugging should occur. Some of this variation may be due to differences in the criteria used by the authors to define slugging, and this should not be surprising given the complex nature of the bubbling-to-slugging transition revealed in this study. Assuming that the trends in

the present MFIX simulations can be experimentally validated, it might be more useful to develop correlations that predict the flows at both the beginning and end of the slugging transition process. This is clearly an area where development of an improved understanding of the physics underlying slugging and a more precise terminology related to its occurrence would be helpful.

Fig. 15 Comparison between this study and slugging correlations

4. Conclusions

Results from the three-dimensional computational simulations of a laboratory-scale fluidized bed indicate that the transition from bubbling-to-slugging is a complex process that occurs over a range of gas flows rather than abruptly at a single flow. The transition process appears to involve a cascade of bubble coalescences that produce size and speed changes which begin near the bed surface and then progress downward toward the distributor as gas flow increases. The state of maximum slugging intensity appears to be reached when the most intense coalescence point approaches the bottom of the bed. The general dynamical trend predicted by our simulations appears to be consistent with the trends observed in other experimental studies of

bubble behavior in viscous liquids and bubbling fluidized beds equipped with bubble imaging capabilities.

Detailed analyses of the simulated bubble patterns and high-speed pressure fluctuations indicate that both amplitude and time-scale statistics derived from the pressure fluctuations can be a useful diagnostic for tracking bubble behavior and the transition from free bubbling to maximum intensity slugging (fully developed slugging). The most useful pressure amplitude statistics include the kurtosis and skewness of pressure fluctuation histograms. Time-lagged autocorrelations in the pressure time series appear to correlate with bubble sizes and speeds. However, the pressure features most useful for diagnosing bubble behavior depend strongly on the vertical location in the bed where the pressure measurements are made. Based on the analyses described here, the optimum location for pressure measurements to monitor bubble behavior related to the slugging transition appears to be near the top of the bed, below the static bed height.

There are several remaining areas that should be investigated concerning the ~~For future~~ bubbling-to-slugging transition in beds of Group B solids:

- Direct comparisons between computational simulations and high-speed experimental void fraction and pressure measurements [73, 74] are needed to validate/improve existing CFD models.
- Computational simulations of the bubbling-to-slugging transition with other CFD approaches such as the Eulerian-Lagrangian Discrete Element Method (DEM) are needed to resolve questions about the best approach for capturing the fundamental physics.
- More detailed studies of the detailed relationship between bubble and pressure dynamics are needed to allow pressure signals to be a useful indicator of bubble patterns.

- Further analyses are needed to understand how spatial voidage and pressure measurement locations can affect the dynamic information available for generating useful diagnostics.
- Computational simulations and corresponding experimental measurements are needed to determine how significant the bubbling-to-slugging transition is likely to be in altering chemical conversion and the efficiency of heat and mass transfer.

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Pressure Statistics

Bubble Statistics