Fluidization in small-scale gas-solid 3D-printed fluidized beds

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Abstract: Additive manufacturing could be used to facilitate the rapid fabrication and testing of small-scale fluidized beds for use in screening applications, such as adsorbent screening for carbon capture. In this work, experiments were performed in order to map the different flow regimes produced in small-scale ($D_h = 3–15$ mm) gas-solid fluidized beds that were fabricated using additive manufacturing using the stereolithography approach. Here, the effects of bed hydraulic diameter ($D_h$), static particle height ($H_s$), and particle type/size ($D_p$ and $\rho_p$) were considered. Pressure drop data and high speed camera images were used to develop simple flow regime maps for these printed beds showing the operating windows for packed bed, bubbling, slugging and turbulence applicable to a wide range of bed size to particle diameter ratios ($D_h/D_p = 20–200$) and gas velocities ($U_g = 1–400$ mm/s) in both ‘2D’ and ‘3D’ bed aspect ratios. Fast Fourier transforms of the pressure drop signals were also used to study the evolution of bubbling/slugging behaviour as the gas velocity was increased by creating 2D colour maps of the frequency spectra. These allowed a new quantitative method to be proposed for the identification of slugging – the point where the dominant frequency in the power spectrum becomes constant as the gas velocity increases. It is concluded in this study that the rougher surfaces generated by additive manufacturing do not influence the fluidization characteristics nor modify the wall effects of small-scale beds. Macro-scale fluidization could also be achieved at smaller $D_h/D_p$ ratios in these 3D printed beds compared to more conventional Plexiglas beds ($D_h/D_p = 75$ compared to $D_h/D_p = 300$).

Keywords: Additive manufacturing, micro-fluidized bed, bubbling, slugging, turbulence

Graphical Abstract:
1 Introduction
Potic et al. (2005) were the first to propose micro-fluidized beds for small-scale screening experiments to realise the benefits of miniaturised chemical reactors: high-throughput testing, low experimental costs, low energy consumption, improved safety, and good heat and mass transfer properties to overcome diffusion-limitations. Further, it has been suggested that micro-fluidized beds also provide better translation of lab scale results to larger scales compared to micro-packed beds, where gradient effects are difficult to translate (Potic et al., 2005). Yu et al. (2010) and Guo et al. (2016) have subsequently developed micro-fluidized bed reactor analysers to study the kinetics of solid reactions (e.g. catalysts). Further examples of the use of micro-fluidized beds for reaction kinetics screening and granulation dynamics are included in the review on solids processing technologies by Wang et al. (2017).

Micro-fluidized beds are distinguished from ‘normal’ fluidized beds according to the influence of surface effects (wall friction) relative to volumetric effects (gravity) (Zivkovic & Biggs, 2015). These ‘wall effects’ typically manifest as delayed fluidization where the higher relative importance of wall friction stabilises the bed as the fluid exerts drag on the particles. Various definitions of micro-scale fluidization behaviour ranging from bed diameters of sub-500 μm (Zivkovic et al., 2013) to 15.5 mm (Guo et al., 2009) have been proposed, though there are several nuances depending on the exact fluidized bed material, particle type and fluid (Zivkovic & Biggs, 2015). Notwithstanding, it has been shown experimentally that the fluidization regime impacts on the kinetics because of mass transfer effects (Jaiboon et al., 2013a). Consequently, numerous groups have researched the flow patterns (Wang et al., 2011; Wang & Fan, 2011; Jaiboon et al., 2013b) and studied the wall effects (Loezos et al., 2002; Liu et al., 2008; Guo et al., 2009; Rao et al., 2010; do Nascimento et al., 2016) in a variety of micro-fluidized beds to understand the fluidization characteristics.

All micro-fluidized beds reported to date have been manufactured by ‘conventional’ processes, and are often constructed from either Plexiglas (PMMA) or quartz tubing and capillaries. Additive manufacturing (AM) is an alternative, yet powerful, fabrication technology that is growing in popularity for the construction of novel chemical reactors and catalysts (Parra-Cabrera et al., 2018), advanced pharmaceutics (e.g. pill delivery) (Pravin & Sudhir, 2018) and advanced heat transfer devices (Jafari & Wits, 2018). AM is a transformative technology, because it removes the rate-limiting step for prototyping, provides a means for realising more complex geometries, and enables separation of the design and fabrication steps allowing for a distributed manufacturing model (where specialist companies can focus solely on providing an additively manufactured object according to a customer’s design).

Thus, there is an obvious potential to exploit AM for the fabrication of novel/advanced fluidized bed designs to improve lab scale screening. This could include designing additional wall structures to promote fluid percolation through the bed to enhance fluid-particle mixing, creation of integrated multi-stage zones, and optimization of the distributor. This latter idea has recently been tested by Odeleye et al. (2018) in liquid fluidized beds. Further, the fast turnaround times between design conception and realisation would allow for rapid optimization of a fluidized bed for any particular application, which would help to find a bed design that avoids any mass-transfer effects.

However, one potential caveat to AM is the surface roughness, which has been shown to affect the mixing in liquid-filled 3D-printed microfluidic channels (MacDonald et al., 2017), which may or may not be advantageous to fluidization. An early study by Zhou et al. (1996) explicitly studied the effect of wall roughness in a circulating fluidized bed by fixing sandpaper to the walls. Here, the protrusions in the sandpaper were around 0.45 mm in size, twice the particle diameter. They found that rough walls produce a flatter local voidage profile across the radial direction of the column with higher voidage at the walls compared to smooth walls. Here, wall roughness had little effect near the centre of the bed, and the authors also showed that there was no statistical difference between the particle descent velocities near the wall region between rough and smooth surfaces. In a later study, it was found that the particle-wall friction factors were expectedly higher when applying sandpaper to the walls compared to smooth walls, but the shape of the trends were preserved (Mabrouk et al., 2008). Several numerical
and experimental studies have further elucidated the influence of wall roughness. Through a ‘two-way coupling’ (where particle motion influences the turbulent gas flow and vice versa), rougher walls increase the fluctuation energies of the particles, which reduces the free path lengths resulting in higher particle-wall collision frequencies and irregular collisions (Lain & Sommerfeld, 2008; Sommerfeld et al., 2004). This increased turbulent dissipation then results in higher particle momentum losses, which increases the slip velocity between the gas and particles resulting in higher pressure drops (Sommerfeld et al., 2004; Kussin & Sommerfeld, 2002).

However, the current published research concerning wall effects and flow regimes in mini and micro-fluidized beds applies mainly to the ‘smooth’ surfaces in which these studies were performed; there is a need to evaluate whether fluidization in an additively manufactured device is feasible. Thus, the present study aims to address this, by studying the fluidization characteristics of Geldart A and Geldart B particles in various sized 3D-printed fluidized beds at a range of static bed heights and across a broad range of gas velocities. Note, the present study is focussed on gas-solid fluidization, which is motivated from a collaborative EPSRC-funded project involving the use of solid adsorbents for capturing CO₂ from industrial processes (McDonough et al., 2018).

2 Methodology
2.1 Fluidized Bed Designs and Additive Manufacturing
Six planar fluidized bed geometries were considered in the present investigation, shown in Figure 1. These designs had hydraulic diameters (Dₕ) ranging from 3–15 mm, giving corresponding gas flow areas of 9–225 mm². Each fluidized bed included a planar distributor plate (5 mm height) containing 1 mm square holes, above which, a 26 μm aperture steel mesh was fitted (using a custom locking pin) to prevent the particles from escaping. Below the distributor was a basic plenum that expanded from a 2 mm diameter inlet tube to the final cross-section width. This plenum was packed with glass wool to avoid maldistribution of the fluidizing air. Each design was manufactured with three sides, which was then sealed with a 3 mm thick Perspex sheet for visualisation. Custom pressure ports were also integrated into each fluidized bed at three different heights to enable the pressure drop to be recorded. These ports were placed 3 mm above/below the distributor plate, and 10 mm beneath the outlet. Each pressure port was also covered with 26 μm aperture steel mesh to prevent particle escape from the bed. Figure 2 shows an example of a 3D-printed fluidized bed with Perspex viewing sheet.

![Figure 1 – CAD models of the planar fluidized bed designs considered for the flow regime mapping experiments | (a) 3x3 mm, (b) 5x5 mm, (c) 5x15 mm (Dₕ = 7.5 mm), (d) 5x25 mm (Dₕ = 8.33 mm), (e) 10x10 mm, and (f) 15x15 mm | the layout of the distributor plates are also shown to scale](image-url)
All small-scale fluidized bed designs were fabricated via additive manufacturing using the stereolithographic (SLA) approach in a Form2 (FormLabs) printer. Stereolithography involves the layer-by-layer construction of parts through the photopolymerisation of a resin. The Form2 used a 405 nm UV laser (140 μm spot size, 250 mW) to cure a translucent methyl acrylate based proprietary resin (FLGPCL02) comprising methyl acrylate monomer (55-75% w/w), methyl acrylate oligomers (35-40% w/w) and photo-initiator additives (10-15% w/w). Construction of each layer involved the laser scanning quickly through the resin tank according to the shape defined by the corresponding slice through the geometry, followed by the build platform being raised by the user-specified Z-axis resolution. The Form2 delivers a maximum XY (horizontal) resolution of 14 μm and Z-axis (vertical) resolutions of 25–100 μm; though 100 μm produced the best surface quality so was used for all designs in this study. A discussion about the elevated wall roughness compared to more standard materials (such as Plexiglas and quartz) is included in Section 3.3.

The procedure for manufacturing parts was as follows: (1) creation of a 3D CAD model of the device using Google SketchUp, (2) conversion of the CAD model to a triangular mesh model (.stl file format), (3) slicing of the triangular mesh model into different layers and conversion of these slices to tool paths for the printer (using PreForm software), (4) 3D printing, and (5) post-processing. Post-processing involved cleaning the parts in an isopropyl alcohol (IPA) ultrasonic bath (FormWash) to remove excess/uncured resin, drying the parts using compressed air, and curing at 60°C for 30–60 min in 405 nm UV light (FormCure). Parts were printed at an angle of approximately 60° from the horizontal in both the X and Y directions to improve resin drainage and improve the stability/quality of the part during printing. Approximately four milli-scale fluidized beds could be manufactured simultaneously in approximately 10 hours, at a total cost of ~£22 of resin.

2.2 Experiment Setup and Procedure

Three differently sized glass microspheres (2.1 g/cm³) and one size of silica particle (2.65 g/cm³) were used in the present investigation. Each of the size distributions was confirmed using a Coulter LS230 sizer. The silica particles had an average size of 93 ± 10 μm (Geldart A), whilst the three glass microsphere samples had average sizes of 82 ± 7 μm (Geldart A), 170 ± 24 μm (Geldart B) and 183 ± 29 μm (Geldart B). Each of the particle sizes were normally distributed, as shown in Figure 3. Three
particle bed heights were considered in each of the six fluidized bed designs, corresponding to dimensional static height ratios of \( H_s/D_h = 2, 3 \) and 4.

**Figure 3** – Particle size distributions of silica particles and glass microspheres obtained from a Coulter LS230 sizer

High-speed videos of the fluidized beds were recorded using a Basler acA1300-200uc camera (169 fps, 1.3 MP) fitted with a COSMICAR Television lens (12.5 mm, 1:14), monitored via Pylon Viewer software. Illumination of the beds was achieved using an LED light panel placed behind the bed and a fiber optic directional lamp (Microlight 150) pointed at the fluidized particles. A Sensirion SDP610 differential pressure transducer was used to simultaneously measure the bed pressure drops. Here, the sensor was connected to two of the pressure ports integrated into the 3D printed design (see Figure 1). The pressure sensor had a working range of ±500 Pa, a precision of 0.001 Pa and 4.6 ms response time. A schematic of the experiment rig is shown in Figure 4 below.

The fluidizing gas in all experiments was compressed air regulated to atmospheric pressure using an AW4000 regulator. This enabled precise control of the volumetric flow rate (3.7–2600 mL/min), which was adjusted using one of four Omega float meters connected in parallel. Experiments were performed as follows. First, the particular fluidized bed was filled with one of the four particle grades to one of the three static height ratios. The particles were then emptied from the bed and weighed to confirm the starting weight before being placed back in the bed (a summary of these weights is included in the supplementary materials document). Pressure drop data and high-speed videos were then recorded for increasing and decreasing air flow rates to ensure any hysteresis effects were captured. Here, 20 s of pressure drop data were recorded along with ~3.5 s of high-speed video (corresponding to 2 GB of data) for each air flow rate (for both the increasing and decreasing gas flow experiments). Due to the size of the video files, videos were only collected for the 93 ± 10 μm silica particles; this was sufficient for visually identifying the flow regimes. The pressure drop across the particle bed was obtained by subtracting the pressure drop measured across the empty bed. It was confirmed that the pressure drop across the glass wool and distributor was at least 50% of the total pressure drop to limit maldistribution of the gas in the bed.

Due to the amount of data collected, only representative examples of the major results have been included in this manuscript. The full set of results are available in a supplementary document, including: pressure drop profiles, snapshots from the high-speed camera videos, animated videos showing representative examples of the flow regimes in each fluidized bed design, and frequency spectra maps.
Figure 4 – Schematic representation of the experiment setup (images of the actual setup are included in the supplementary materials document)
3 Results and Discussions
3.1 Visual Observation of Flow Regimes
Figure 5 shows representative examples of the main flow regimes observed in the six 3D printed fluidized bed designs considered in the present study. Accompanying descriptions of these regimes are included in the following bullet point list, which are largely similar to the observations previously reported by Wang et al (2011) for smooth tubes. Animated videos of each flow regime from Figure 5 along with accompanying snapshots from all bed designs at different gas velocities are included in the supplementary materials document. Figure 6 then shows snapshots from the high-speed videos showing the progressions of the flow regimes as the superficial gas velocity was increased in two of the bed designs. These regimes were generally the same for both the Geldart A and B particles considered (Figure 3), with the main differences discussed in Section 3.2.2.

- **Packed bed.** Fixed particle bed structure and an almost linear increase of the bed pressure drop with increasing gas velocity. For Geldart A particles, the pressure drop was very stable, indicating no internal movement of the particles and no gas bubble formation. For Geldart B particles, the pressure drop did slightly fluctuate prior to the minimum fluidization point, indicating the formation of gas bubbles prior to fully support of the bed
- **Minimum fluidization.** Slight bed expansion was observed along with a corresponding slight decrease in pressure drop (a consequence of wall friction, explained later). The pressure drop remained relatively stable with further increases in the gas velocity
- **Particulate fluidization.** Following minimum fluidization, there was little motion of the particles across the bulk of the cross-section, though some small bubbles were observed at the walls. Additionally, very small gas bubbles were observed close to the distributor plate, but these quickly collapsed into the particle phase
- **Bubbling.** With a further increase in the gas velocity, consistent bubble formation was observed across the whole distributor. Bubbles rose up to the surface across the full cross-section of the bed and the mixing was qualitatively good. There was also minimal variation in the particle bed height
- **Slugging.** Large gas slugs developed across the majority of the cross-section of the bed due to bubble coalescence. Large variation in the particle bed height occurred as a result of continuous geyser eruptions occurring at the free surface
- **Turbulence.** Separate gas bubble/slug and emulsion phases were no longer visible. The majority of the cross-section of the bed became a ‘homogeneous’ blend of particles within the gas. The particle bed density decreased from the distributor towards the free surface, and the bed height remained relatively constant (compared to slugging)

![Figure 5 – Representative examples of different flow regimes produced in different fluidized bed designs (animated .gif versions of these figures are included in the supplementary materials)](image-url)
In this study, the gas velocities were not high enough to observe fast fluidization, which is the regime where entrainment of the gas particles occurs allowing for their circulation. Instead, a minor form of elutriation was observed throughout the experiments were a small number of particles would statically adhere to the Perspex viewing window. This resulted in a slight decrease of the pressure drop across the bed as the gas velocity was increased which was irreversible within a particular experiment. This resulted in a small hysterisis of $U_{mf}$ in the fluidization and defluidization experiments (increasing and decreasing gas flow respectively). However, it was not possible to quantify this elutriation phenomenon using high-speed camera data; the effect is observable in Figures 7a and 7b, where the pressure drops were slightly smaller during the decreasing gas flow experiments compared to increasing gas flow experiments.

Norouzi et al. (2011) found that in shallow beds ($H_s/D_h = 0.5$), the gas bubbles mainly rise up near the walls with the solids returning to the distributor plate predominately at the centre of the column. For $H_s/D_h = 1$, this flow pattern is reversed and for $H_s/D_h = 1.5$, a hybrid is formed with solids flowing downward in the central region at the bottom of the bed, and solids flowing upward in the central region at the top of the bed. In the present study, this circulation of the solids within the bed was difficult to quantify.
3.2 Pressure Drop Profiles

3.2.1 Effect of Gas Superficial Velocity

Figure 7 – Pressure drops across the 93 ± 10 μm silica particles at different superficial gas velocities in different fluidized bed designs | stainless steel mesh distributor | (a) 3x3 mm, (b) 5x5 mm, (c) 5x15 mm ($D_h = 7.5$ mm), (d) 5x25 mm ($D_h = 8.33$ mm), (e) 10x10 mm, and (f) 15x15 mm | filled symbols refer to increasing gas velocity & empty symbols refer to decreasing gas velocity | dashed lines refer
to the theoretical pressure drops based on the static bed weights (equation 2) | the pressure drop profiles obtained with the glass beads are included in the supplementary materials document

Figure 7 shows the effect of gas superficial velocity on the pressure drop recorded across the 93 ± 10 μm silica particles in each of the six fluidized bed designs considered for this study (refer to Figure 1). Initially, the pressure drops across the bed increased as the gas superficial velocity increased. Here, there was a slight parabolic trend (more apparent in Figure 7a) whose effect is captured by the friction term in the Ergun equation (equation 1). Prior to the minimum fluidization point there was a small pressure overshoot, a consequence of wall effects, after which, the pressure drop remained approximately constant as the gas velocity was increased further. Although the magnitude of the pressure overshoot increased with increasing bed size, when this overshoot was normalised against the bed volume, the pressure overshoot effect was observed to decrease with increasing bed diameter. Further discussion is provided in Section 3.3.

\[
\frac{\Delta P_{\text{Ergun}}}{H_s} = 150 \left( \frac{1 - \varepsilon}{\varepsilon^3} \right)^2 \frac{\mu g U_g^2}{\phi^2 D_p^2} + 1.75 \frac{1 - \varepsilon}{\varepsilon^3} \frac{\rho_g U_g^2}{\phi D_p}
\]

The theoretical pressure drops are indicated in Figure 7 by the dashed lines, and were calculated by considering the weight of the particles according to equation 2 (Guo et al., 2009). Here, \( H_s \) is the static height of the particle bed, \( \rho_p \) is the particle density, \( \rho_g \) is the gas density, \( g \) is the gravitational acceleration constant and \( \varepsilon_m \) is the voidage at the minimum fluidization point. The value of \( \varepsilon_m \) was itself calculated from equation 3, where \( V_{mf} \) is the total bed volume at the minimum fluidization point, \( V_p \) is the total particle volume, \( m \) is the mass of the particle bed (measured before each experiment), \( A \) is the cross-sectional area of the fluidized bed and \( H_{mf} \) is the height of the particle bed at the minimum fluidization point (taken from the high speed videos).

\[
\frac{\Delta P}{H_s} = [\rho_p (1 - \varepsilon_{mf}) + \rho_g \varepsilon_{mf}] g
\]

\[
\varepsilon_{mf} = \frac{V_{mf} - V_p}{V_{mf}} = \frac{H_{mf} A - m}{\rho_p H_{mf} A}
\]

For the 3x3 mm, 5x5 mm, 5x15 mm and 5x25 mm fluidized bed designs, it can be seen in Figure 7 that the pressure drops measured during the fluidization experiments were equal to the predicted pressure drops based on the calculated buoyant weight (equation 2). However, for the 10x10 mm and 15x15 mm designs, the pressure drops measured during fluidization were significantly lower than the predicted pressure drops, around 30–38% and 15% respectively.

Previous studies have also observed differences between the measured and predicted pressure drops which have been attributed to wall effects (Guo et al., 2009; Sánchez-Delgado et al., 2011). Here, the enhanced wall friction due to larger surface area to volume ratio of smaller fluidized beds contributes to the support of the particles, producing a smaller weight force that needs to be balanced by the upward gas flow. I.e. smaller beds produce a larger offset from the predicted pressure drop. The transition from micro to macroscale behaviour has subsequently been identified using this principle; for example, \( D_h \geq 15.5 \text{ mm} \) marked the transition to macroscopic characteristics for Plexiglas tubes containing fluid catalytic crack (FCC) particles and quartz sand with diameters of 30–83 μm (Guo et al., 2009). In contrast, in the present study it was the larger fluidized bed designs where an offset between the measured and predicted values was observed.

Tsinontides & Jackson (1993) also observed an offset between the measured and theoretical pressure drops in larger diameter beds, whilst Loezos et al. (2002) and Srivastava & Sundaresan (2002) additionally found that the pressure drop offset increased as the bed diameter was increased. For example, using 88 μm FCC particles, the percentage offset reportedly increased from 2.0% to 4.5% as the tube diameter was increased from 0.5 in to 2.0 in (Loezos et al., 2002). The occurrence of a pressure drop offset implies that the beds are not in a completely fluidized state, with either the distributor and/or walls contributing to the support of the particles. Loezos et al., 2002 attributed the offsets to partial...
support of the particles by the distributor, where some particles fall back down to and then bounce on the distributor following the passage of a bubble. Alternatively, Srivastava & Sundaresan (2002) similarly reasoned that the offset cannot be due to wall friction alone (because the offset grows as the diameter increases), instead conjecturing that larger beds present more pronounced lateral inhomogeneity in the bulk density, which presumably might influence gas motion. This idea is supported by the observations of Sánchez-Delgado et al. (2011), who studied the minimum fluidization velocities in 2D (high aspect ratio) beds. They observed that gas bubbles preferentially percolate through the central region of the bed because of bubble coalescence, resulting in non-uniform voidage through the bed. Thus, for non-uniform gas distribution in the bed, Sánchez-Delgado et al.’s (2011) observations suggest that the wall regions might be additionally supported through wall friction and/or additional electrostatic cohesion forces.

For the 10x10 mm and 15x15 mm designs, it is likely that the deviations between the measured and predicted pressure drops were a consequence of the distributor plate partially supporting the weight of the particle bed, where the stainless steel mesh placed over the 1x1 mm holes in the distributor plate were not sufficient to avoid gas channeling in the lower parts of the bed. I.e., the particles in the lower part of the bed between these 1x1 mm holes in the distributor plate were not in a fully fluidized state.

To validate this hypothesis, two new sets of 10x10 mm and 15x15 mm fluidized beds were fabricated. One set repeated the stainless steel mesh design shown in Figure 2 whilst the second replaced the stainless steel mesh with a 1.5 mm thick porous PTFE plate with 21 μm pore size (SPC Technologies Ltd., UK). Figure 8 compares the pressure drop profiles of the original and repeated stainless steel mesh designs with the porous PTFE plate design for the 93 μm silica particles using $H_s/D_h = 2$. Here the black dotted line indicates the average ‘weight force’ of the three particle beds (determined using equation 2). It can be seen that both mesh designs produced pressure drops below the theoretical value, whilst the pressure drop recorded using the porous plate matched the theoretical pressure drop in both the 10x10 mm and 15x15 mm designs. This confirms that wall friction had minimal influence over the fluidization behaviour and that the air leakage did not account for the deviation in measured pressure drops, meaning that the distributor using the stainless steel mesh indeed supported only part of the particle bed.

Based on the results in Figure 7 and the results of the porous plate distributor design in Figure 8, it is concluded that the increased wall roughness generated through additive manufacturing does not adversely affect the fluidization process in small-scale beds (further discussed in Section 3.3). It should be noted here that the stainless steel mesh was primarily used to prevent particle seepage in the distributor plate orifices, and was itself not the distributor. The original aim was to have an entirely 3D-printed fluidized bed with 3D-printed orifice plate distributor, where one could easily 3D-print two parts...
with the stainless steel mesh sandwiched between them, eliminating the need for an external porous plate. Although the magnitude of the pressure drops were different between the stainless steel mesh and porous PTFE plate, the fluidization characteristics themselves were the same; both configurations produced the same minimum fluidization velocity, slugging onset velocity and critical velocity for turbulence (see supplementary data).

### 3.2.2 Effect of Particle Diameter

Figure 9 shows the pressure drops produced by the four different particles in the 5x5 mm and 15x15 mm fluidized bed designs (Figure 1). Here, the theoretical pressure drops based on equation 2 for the silica particles have been included. All four particles investigated (Figure 3) could be fluidized in each bed design. The A and B particle types produced slightly different behaviours in the packed bed regime, with the larger Geldart B particles producing a shallower gradient as the gas velocity was increased. This is explained by considering the Ergun equation (equation 1), which contains contributions to the pressure drop from frictional losses and the turbulence of the fluid (via a modified Reynolds number). Larger particle diameters increase the void spacing resulting in reduced frictional losses.

Following the minimum fluidization point in the 5x5 mm bed design, the larger particles produced a smaller pressure drop. This is because the particle size relative to the bed size was larger (i.e. \(D_s/D_h\) was smaller), meaning wall effects were more significant. This is quantified by the voidage. For the Geldart A and B particles in the 5x5 mm design, the average bed voidages at the minimum fluidization point were \(\epsilon_{mf} = 0.44\) and \(\epsilon_{mf} = 0.37\) respectively. The larger average value of \(\epsilon_{mf}\) for the Geldart A particles indicates that they were not able to pack as closely together because of the increased relative contribution of wall friction helping to support the particle bed. In the 15x15 mm design, more comparable pressure drops for each of the particle types were observed once fluidization initiated, suggesting that wall effects were negligible in this design.

The other significant difference observed in the present study was the apparent onset of bubbling prior to the minimum fluidization point when using the Geldart B particles. This was indicated by the standard deviation of the pressure drop signal becoming non-zero whilst the bed was still in the packed bed state. These results are further discussed in Section 2.6.2.

### 3.3 Additive Manufacturing and Wall Effects

Wall effects become more significant as the diameter of the bed (\(D_h\)) decreases because the contact area between the bed and the walls is proportional to \(D_h^2\), whereas the volume is proportional to \(D_h^3\). Thus, at a critical bed diameter, the relative magnitude of the frictional force between the bed and the wall
(dependent on the bed-wall contact area) exceeds the magnitude of the drag force exerted on the particles by the fluidizing gas. This friction both stabilises the bed, delaying the onset of fluidization, and contributes to the support of the particles, sometimes increasing the deviation between measured and theoretical pressure drop (Guo et al., 2009). The friction between the particle bed and wall arises because of adhesive forces (such as Van der Waals forces), surface tension effects and static cohesive effects (Zivkovic & Biggs, 2015; Sánchez-Delgado et al., 2011). Increasing the roughness of a surface will increase the contact area, which will likely influence the friction (Wilson et al., 2017; Mabrouk et al., 2008).

Depending on the orientation of the surface, the layering process during additive manufacturing combined with the finite resolution of the particular printing technology can lead to a “stair stepping effect” which increases macro-scale roughness at the surface. MacDonald et al. (2017) compared the effects of surface roughness on the mixing rate in a microfluidic chip using three 3D printing technologies: Fused Deposition Modelling (FDM), Polyjet and Digital Light Processing Stereolithography (DLP-SLA). Their DLP-SLA printer produced a surface roughness of just 0.35 μm whereas the FDM printer produced a surface roughness of 10.97 μm. Thus, in laminar flow tests (at flow rates of 25–100 μL/min) they found that the extent of mixing was significantly lower in the DLP-SLA device in a 750×500 μm channel compared to the FDM technology.

The transition from macro-scale to micro-scale fluidization behaviours defines the point where wall effects become significant. Intuitively, the surface roughness might influence this transition point similar to how mixing in microfluidic channels are affected (MacDonald et al., 2017). Okafor et al. (2017) determined that the root mean square surface roughness of the Form2 printer used in the present study is 2.9 μm (using the 100 μm layer height setting). For reference, the average surface roughness of conventional micro-fluidized bed materials are reportedly: <0.2 μm for polished PMMA (Kuhar & Funduk, 2005), and <5 nm for polished quartz (Bo et al., 2014). Figure 10 shows three methods that were used to identify the transition between macro- and micro-scale behaviour for all experiment configurations considered in this study.
Figure 10 – Transition points between micro- and macro-scale fluidization behaviour with stainless steel mesh distributor. (a) $U_{mf}$ vs $D_h/D_p$, (b) Comparison of measured $U_{mf}$ with predicted $U_{mf}$ (equation 4), and (c) pressure overshoot to bed volume ratio vs $D_h/D_p$.

Figure 10a shows a plot of the measured minimum fluidization velocities against the ratio of bed hydraulic diameter to particle diameter ($D_h/D_p$). As the bed diameter decreases, higher gas velocities are required to overcome the increasingly significant influence of wall friction (Rao et al., 2010; Liu et al., 2008; Loezos et al., 2002). In Figure 10a, the $U_{mf}$ values decreased exponentially with increasing $D_h/D_p$, similar to observations of Guo et al. (2009), tapering around $D_h/D_p = 75$. The effect of $H_s/D_h$ is also more apparent at smaller $D_h/D_p$ ratios, where taller beds produced a higher minimum fluidization velocity, which also agrees with the observations of Guo et al. (2009).

The Geldart equation can be used to predict the pressure drops of macroscale fluidized beds (Geldart, 1973). Figure 10b shows the experimentally measured minimum fluidization velocities vs the predicted values using equation 4. As the bed hydraulic diameter increased, the predicted values approached the experimentally measured values. However, for the smaller beds (3x3 mm and 5x5 mm designs), equation 4 massively underpredicted the minimum fluidization velocities because this model does not capture wall effects. Guo et al. (2009) presented a modified equation for predicting $U_{mf}$ that also contained the bed diameter and static bed height terms to account for wall effects, though this model was derived outside of the range of $D_h/D_p$ considered in this study.

$$U_{mf} = \frac{8 \times 10^{-4} g D_p^2 (\rho_p - \rho_g)}{\mu_g}$$  \hspace{1cm} (4)
During fluidization onset (when increasing the gas velocity), pressure overshoots are often observed when wall effects are significant. The term \( \Delta P_A/\text{mg} - 1 \) has been proposed for describing this pressure overshoot (Loezos et al., 2002). However, Liu et al. (2008) argue that this is ambiguous, because the bed is not in a fluidized state during the overshoot, meaning the full weight of the bed (described by mg/A) is not being supported by the gas. Instead, they proposed the parameter \( \Delta P_{mf}/V \), where \( \Delta P_{mf} \) is the difference between the maximum overshoot pressure drop and the pressure drop during fluidization and \( V \) is the volume of the bed. This parameter represents the extra pressure drop required per unit volume of material to overcome frictional effects. Figure 10c shows \( \Delta P_{mf}/V \) plotted against the ratio of bed hydraulic diameter to particle diameter \( (D_b/D_p) \). There is a general trend of decreasing \( \Delta P_{mf}/V \) with increasing \( D_b/D_p \) and there is no obvious effect of \( H_s \), which is in agreement with the definition proposed by Liu et al. (2008). However, prior to \( D_b/D_p \sim 75 \) the data were quite spread, which could be a consequence of differing particle-wall static cohesion and uneven voidage distribution effects.

Wall effects refer to the increasing relative importance of friction as the surface area to volume ratio increases in smaller scale fluidized beds. The enhanced friction of these smaller beds increases the magnitude of the pressure overshoot and increases the minimum fluidization velocity because the higher relative importance of friction stabilises any movement of the bed (Loezos et al., 2002). The transition from micro to macro scale fluidization behaviour can consequently be defined as the points where \( U_{mf} \) and \( \Delta P_{mf}/V \) become constant as \( D_b/D_p \) is increased, which indicate the points where wall effects become negligible. For the 3D printed fluidized beds considered in this study containing glass and silica particles, both of these transitions occurred at \( D_b/D_p = 75 \) for all particles and static bed heights studied.

Rao et al. (2010) extended the Ergun equation for the prediction of \( U_{mf} \) whilst accounting for wall effects in small scale fluidized beds as shown in equations 5, 6 & 7. Here, the model was derived for various acrylic beds containing glass beads (\( \rho_p = 2.5 \text{ g/cm}^3 \)) and polystyrene particles (\( \rho_p = 1.25 \text{ g/cm}^3 \)) with particle diameters of 105–600 \( \mu \text{m} \). The constants \( k_1 \) and \( k_2 \) were found to be 610 and 30.1 respectively through curve fitting. In these equations, \( \varepsilon \) is the voidage, \( \phi \) is the particle’s ‘friction angle’ and \( \phi \) is the particle sphericity.

\[
\begin{align*}
\left[ 1.75 \frac{1 - \varepsilon}{\varepsilon^3} - 4k_2 \tan \varphi \left( \frac{\phi D_p}{D_t} \right) \left( \frac{H_s}{D_h} \right) \right] Re^2 + \left[ 150 \frac{(1 - \varepsilon)^2}{\varepsilon^3} - 4k_1 \tan \varphi \left( \frac{\phi D_p}{D_t} \right) \left( \frac{H_s}{D_h} \right) \right] Re = (1 - \varepsilon)Ar \\
Re = \frac{\rho_g (\phi D_p) U_{mf}}{\mu_g} \\
Ar = \frac{(\phi D_p)^3 (\rho_p - \rho_g) \rho_g g}{\mu_g^2}
\end{align*}
\]

By applying these equations to the silica and glass particles of the present study, the elbow in the \( U_{mf} \) vs \( D_b/D_p \) plot is predicted to occur around \( D_b/D_p = 200–300 \), similar to the transition point reported by Guo et al. (2009) for a Plexiglas column containing 51 \( \mu \text{m} \) FCC particles: \( D_b/D_p = 300 \). The predicted \( U_{mf} \) from this model under macroscopic fluidization conditions was \( \sim 16 \text{ mm/s} \), which is comparable to the value of 20 mm/s shown in Figure 10a. Thus, although the additively manufactured surfaces have a rougher surface in comparison to the polished PMMA surfaces (i.e. acrylic & Plexiglas), macro-scale fluidization could be achieved at smaller \( D_b/D_p \) ratios.

Using the two-fluid modelling approach, it has been shown that wall friction does influence the fluidization hydrodynamics (Li & Benyahia, 2012; Johnson & Jackson, 1987). In particular, Khan & Shamim (2017) found that higher wall roughness (modelled by increasing the specularity coefficient in the Johnson and Jackson boundary condition) produced smaller average bubble sizes, suggesting that slugging is somewhat suppressed. This might explain why macroscopic fluidization characteristics could be observed at lower \( D_b/D_p \) ratios using the rougher additively manufactured beds.
Another explanation for the difference between 3D-printed and PMMA beds is the PMMA surface might generate greater electrostatic cohesion between the particle bed and the wall compared to the cross-linked PMA material created using the Form2 printer. Electrostatic effects are complex phenomena that are difficult to predict. Glass particles are known to accumulate more electrostatic charge than plastic ones (Mehrani et al., 2005), and electrostatic charges increase as the bed height increases (Rojo et al., 1986) and as the particle size and gas velocity increase (Guardiola et al., 1996). Sánchez-Delgado et al. (2011) consequently suggests that electrostatic effects should be included in the momentum balance; these effects may be captured in the values of the two coefficients (k_1 and k_2) in equation 5. Others researchers have simply grounded the walls to prevent charge build up altogether (Saxena & Jadov, 1983). In the present study, a small number of particles were qualitatively observed to adhere to the Perspex viewing window, but it could not be visually confirmed if the particles also adhered to the printed polymer surfaces in the same manner.

Irrespective of the cause, the 3D printed polymer was able to fluidize particles with minimal wall effects at smaller D_h/D_p ratios than conventional manufactured materials. It should be noted that the mean roughness of the AM-surfaces were around one order of magnitude smaller than the particle diameters used in this study. For situations where the magnitude of the surface roughness is at a similar scale to the particle diameter (e.g. using FDM printing technologies or finer particles), the wall roughness could be more significant, as recognised in the earlier results of Zhou et al. (1996) and Mabrouk et al. (2008).

3.4 Identification of Flow Regime Transitions

3.4.1 Minimum Fluidization, Minimum Bubbling and Turbulence

The interception point between the pressure drop profiles extrapolated from the packed bed and fluidization regimes during defluidization experiments defines the minimum fluidization velocity. The bubbling and turbulence regimes were also quantitatively determined by considering the standard deviation of the pressure signal. Figure 11 shows the standard deviations of the differential pressure fluctuations measured in each of the six fluidized bed designs containing the 93 ± 10 μm silica particles as a function of the gas velocity (U_g).

The onset of bubbling fluidization (U_{mb}) can be simply visually identified as the point where gas bubbles start erupting from the surface of the particle bed (Kong et al., 2017). More quantitatively, bubbling fluidization can be described as the point where pressure fluctuations start to grow from zero (Leu & Tsai, 2009). With a pressure sensor placed in the plenum and a second sensor placed above the distributor (the configuration used in this study; Figure 4b), Leu & Tsai (2009) note that the U_{mb} increases as the separation distance of the probes increases. This is attributed to attenuation of the pressure signal; i.e. the sensitivity to the smallest bubble eruptions is lost. However, the values of U_{mb} were the same as U_{mf} for the Geldart A particles used in this study, showing that the present experiment configuration was sensitive enough to capture all gas bubble scales. With Geldart B particles, U_{mb} was smaller than U_{mf} in some of the experiment configurations, shown later in Figures 16b, 16d and 16f.
Figure 11 – Standard deviations of the pressure drop fluctuations as a function of gas superficial velocity in different fluidized bed designs | stainless steel mesh distributor | 93 ± 10 μm silica particles | (a) 3x3 mm, (b) 5x5 mm, (c) 5x15 mm (Dh = 7.5 mm), (d) 5x25 mm (Dh = 8.33 mm), (e) 10x10 mm, and (f) 15x15 mm | filled symbols refer to increasing gas velocity & empty symbols refer to decreasing gas velocity

Numerous methods have been proposed to identify the critical velocity (Uc) for the onset of turbulence with varying degrees of success, including: visualisation, bed expansion, voidage fluctuations and pressure fluctuations (reviewed by Bi et al. (2000)). Wang et al. (2011) found that the bed expansion, voidage fluctuation and pressure fluctuation methods predicted different values of Uc, ranging from 0.04–0.10 m/s in a simulated 1.4x3 mm cross-sectional domain containing 75 μm diameter particles.
Zhu & Zhu (2008) also observed differences between the solids concentration and pressure fluctuations methods. They attributed this to the different response dynamics of the gas and solid phases; the gas phase more rapidly transitions to turbulence and mainly affects the pressure signal, whilst the denser solid phase (described by the solids concentration) responds more gradually. Measurement of the pressure fluctuations are generally simpler to implement and is the most popular approach used in the literature (Bi et al., 2000; Wang et al., 2011). First described by Yerushalmi & Cankurt (1979), here, \( U_t \) is defined as the point where the maximum pressure fluctuation occurs as the gas velocity is increased. This is because turbulence is defined by a loss of distinction between continuous and dispersed phases (Bi et al., 2000); there are no distinct bubbling features so the gas emerges from the top of the particle bed more consistently, reducing the signal variation.

As shown in Figure 11, the onset of turbulence could be clearly identified in the 3x3 mm, 5x5 mm and 5x15 mm designs. Three different types of transition to turbulence have been described: (i) Type 1: instant transition from bubbling to turbulence if slugs can’t form \( \left( \frac{d_{\text{g,max}}}{D_t} < 0.7 \right) \), (ii) Type 2: gradual transition where slugging structures slowly disappear, and (iii) Type 3: gas jet penetration occurring in shallow beds \( \left( \frac{H}{D_t} < 2 \right) \) where slugs have insufficient room for development (Bi et al., 2000). Based on the results in Figure 11, it can be seen that the transition mainly exhibits the Type 2 characteristic, where slugging gradually subsides as the bubble and emulsion phases become less distinct and bubble break-up dominates bubble coalescence. In the 5x25 mm and 10x10 mm designs with \( \frac{H}{D_h} = 2 \) (Figures 11d and 11e), it can be seen that the standard deviation did not reach a peak, showing that turbulence was not attained within these experiments.

### 3.4.2 Slugging

Gas slugs develop from the coalescence of bubbles as they move up through the bed. Though if the bed is large enough, then a maximum stable bubble size might be attained where an equilibrium is reached between coalescence and break-up (Shen et al., 2004; Kong et al., 2017). Additionally, for slugs to form the bed height aspect ratio should be at least \( \frac{H}{D_t} \geq 2 \) (Agu et al., 2017). Two definitions of slugging have been proposed in the literature. Wang & Fan (2011) define slugging as the point when the bubble size becomes comparable to the bed size. Whereas, Kong et al. (2017) observed two types of slugs: axi-symmetric slugs (evenly spread across the full cross-section of the column) and wall slugs. The latter exhibited similar unfavourable geyser and reduced particle-gas contact as the former even though the slugs were only in contact with one of the walls. They subsequently used a more relaxed bubble size of 0.66D to denote the onset of slugging.

Bubbling and slugging must be treated as independent phenomena because heat transfer and reaction conversions are negatively affected in the slugging regime (Kong et al., 2017). Jaiboon et al. (2013a) confirmed this experimentally, showing that slugging had a detrimental impact on the CO\(_2\) uptake into K\(_2\)CO\(_3\) solid sorbents compared to the turbulent and “multi-bubbling” regimes.

Broadhurst & Becker (1975) qualitatively visualised a fluidized bed to detect the onset of slugging, defining a slug as having a continuous ‘floor’ around the circumference of the bed (similar to Wang & Fan (2011)). Leva et al. (1951) alternatively proposed that slugging occurs when the pressure fluctuations are around 5–10% of the average pressure drop, matching the definition of slugging proposed by Kong et al. (2017). Ho et al. (1983) later presented a more quantitative approach, defining the onset of slugging as the point where the bubble rise velocity dips slightly before continuing to increase as the gas velocity is increased. The bubble rise velocity can be determined by cross-correlating two pressure fluctuation signals measured in the bed. This rise velocity dip occurs because when the gas bubbles coalesce sufficiently to fill the column, extra drag is exerted on the slug by the walls slowing its ascent (Fan et al., 1983); further increases in the gas velocity then overcome this drag, restoring the continued increase of the rise velocities. Baeyens & Geldart (1974) and Dimattia et al. (1997) identified incipient slugging as the point when the bubbling frequency (taken from the pressure fluctuations) becomes independent of the gas velocity. This method is formalised by Noordergraaf et al. (1987) and Jainboon et al. (2013b), who propose the assessment of the frequency domains of the measured pressure fluctuations. Under slugging conditions, a single predominant frequency tends to be produced because the slugs are uniform and less variable than individual gas bubbles. However, Noordergraaf et al. (1987)
also note that this method might not be suitable for fine particles if the pressure fluctuations are below the sensitivity of the pressure sensor. One of the latest state-of-the-art approaches proposes the measurement of the solids fraction fluctuation in two cross-sectional planes within the bed (Agu et al., 2017). The difference between the standard deviations of the upper and lower planes undergoes a maximum at the onset of slugging, because increased gas velocities create more gas bubbles at the lower plane (increasing the solids fraction fluctuation) and greater coalescence at the upper plane (decreasing the solids fraction fluctuation).

Figure 12 – Examples of raw pressure drop signals and their corresponding frequency spectra recorded in different flow regimes in the 5x5 mm fluidized bed containing 93 ± 10 μm silica particles | stainless steel mesh distributor | H/Dₕ = 2 | (a) packed bed regime (Uₑ = 27.3 mm/s), (b) bubbling regime (Uₑ = 57.7 mm/s), (c) slugging regime (Uₑ = 239.3 mm/s) and (d) turbulent regime (Uₑ = 345.3 mm/s) | (e) corresponding frequency spectra for (a)–(d)

In the present study, the frequency domains could be easily computed using the measured differential pressures by performing a fast Fourier transform. Figure 12 shows representative examples of the time domain pressure drop signals and their corresponding frequency spectra produced in four different flow regimes observed in the 5x5 mm fluidized bed design containing Geldart A particles (93 ± 10 μm silica). Here, the fluidization regimes were identified according to visual inspection of the high speed camera images using the definition of slugging proposed by Kong et al. (2017).

In the packed bed regime (Figure 12a), the time domain signal was stable because there was no movement of the particle bed; the particle adhesion forces were greater than the convective forces resulting in a featureless frequency spectrum. Following the minimum fluidization velocity, bubbling was then observed where minor fluctuations of the pressure drop were observed in the time domain signal (Figure 12b). These fluctuations occurred because of the continual eruption of small gas bubbles from the surface of the particle bed. The corresponding frequency spectrum was broad, and did not show any obvious dominant frequencies, which can be interpreted as a range of bubble sizes/time scales being produced. Following bubbling, slugging behaviour occurred which was characterised by large periodic fluctuations of the pressure drop (Figure 12c). Although the frequency spectra was still broad,
the distribution shows a more obvious Gaussian shape with the distribution centred around 11–12 Hz, suggesting that the gas slugs were more uniform in size than the bubbles in the preceding regime. This is because a narrower range of gas bubble sizes fits the definition of slugging; the slug size is limited by the diameter of the bed, resulting in the slugs forming at predominately the same time scales compared to the bubbling regime. Finally, with further increases of the gas velocity the turbulent regime was observed. As observed in previous studies (Wang et al., 2011; Bi et al., 2000), and in the high speed camera results (Figure 5), there was a loss of distinction between bubbling and emulsion phases. Here, a single ‘homogenised’ gas-solid regime with decreasing density was observed when moving away from the distributor. Consequently, less variation was observed in the time domain pressure drop signal (Figure 12d) because of the decreased gas bubble emergence rate from the bed. Similar to the bubbling regime, the resultant frequency spectra was broader and showed no obvious dominant frequencies in comparison to the slugging regime.

The frequency spectra produced within the slugging regimes of different sized fluidized beds are shown in Figure 13. Here, the distributions have been normalised and offset on the y-axis to allow for easier comparison. It can be seen that as the hydraulic diameter of the bed was increased, the distributions associated with slugging behaviour became broader. This is most likely because the smaller beds imposed a more stringent size limit of the gas bubbles/slugs. This is exemplified in the results of the 3x3 mm design, where a well-defined and narrow peak centred around 10.3 Hz was observed in the frequency spectrum; a result of the gas slugs all being of equivalent size and therefore being produced on similar time scales (characterised by a common time constant). Although the larger beds presented broader frequency spectra, interestingly several dominant peaks were also observable, especially in the 10x10 mm design. This implies that although a broad range of gas slug sizes can produce the slug flow response (bubble sizes ranging from around 0.66Dh–Dh), some slug sizes/time scales are naturally more stable than others. These favoured/stable frequencies are likely to be a function of the distributor design, and warrants further investigation to understand this phenomenon.

Figure 13 – Frequency spectra produced in the slugging regime in different fluidized bed designs using the stainless steel mesh distributor
3.4.3 Slugging Onset Identification using Frequency Spectra Maps

To identify slugging in the present study, a hybrid of the frequency spectra methods of Noordergraaf et al. (1987) and Jainboon et al. (2013b) and the stagnating bubbling frequencies of Baeyens & Geldart (1974) and Dimattia et al. (1997) was adopted. Figure 14 shows three sets of colourmaps of the frequency spectra, which were subsequently obtained by cubically interpolating the frequency spectra recorded at each gas flow rate across the full gas flow rate range considered. Here, the colour intensity has been normalised between 0 and 1 for each gas flow rate; deep blue indicates low intensity (low frequency dominance) while yellow shows high intensities (high frequency dominance). The three maps correspond to the data obtained using the 5x5 mm, 10x10 mm and 15x15 mm fluidized beds containing 93 ± 10 μm silica particles. The gas superficial velocity has also been normalised against the minimum fluidization velocity so that the x-axis shows the fluidization number. For the 5x5 mm and 10x10 mm fluidized bed designs, the respective limits of $U_g/U_{mf} \sim 6$ and $\sim 11$ corresponded to maximum expansion of the particle bed within the height available, whilst for the 15x15 mm design, the limit of $U_g/U_{mf} \sim 5$ corresponded with a limit of the gas superficial velocities that could be applied with the flowmeter setup shown in Figure 4.

The minimum fluidization, bubbling and turbulent boundaries identified from the methods described in Section 3.4.1 have been superimposed onto Figure 14. In all bed designs, the packed bed regime was clearly identified by no structure in the frequency domain, presenting a constant deep blue colour in the maps shown in Figure 14. As already described, this was because there was no development of bubbling structures within the stationary bed. Following the onset of fluidization, small fluctuations in the pressure drop were then observed (Figure 12b), which manifested as a broad frequency spectrum. In the 5x5 mm design, as the gas velocity was increased, the dominant frequency (intense yellow colour) increased before quickly reaching a constant value. The evolution of the dominant frequency with increasing gas velocity has been highlighted with the black trend lines.

As discussed by Baeyens & Geldart (1974), the point where the dominant frequency plateaued was taken as the onset point of slugging. However, in both the 10x10 mm and 15x15 mm designs, the dominant frequency underwent a maximum before decreasing to a stable value. This range of gas flow rates is taken as a transitional regime where slugs begin to experience extra drag from the walls. This could correspond to the zone where the gas slug diameters are $0.66D_t - D_t$, as proposed by Kong et al. (2017). The dominant frequencies in the slugging regimes in this study were around 5–12 Hz, with smaller frequencies being produced using larger $H_s/D_t$ ratios (see supplementary data). Vakhshouri & Grace (2010) also observed that the bubbling frequency is dependent on the static particle height, and proposed equation 8. Interestingly, this equation predicts a similar bubbling frequency range of 5.0–12.9 Hz for static bed heights ranging from 40 mm (largest in this study) to 6 mm (smallest in this study). Vakhshouri & Grace (2010) also noted that both the plenum volume and distributor plate design influence the bubbling frequency; slightly higher frequencies (~10%) were reported for a multi-orifice distributor plate when reducing the plenum volume from around 0.42 to 0.005 m$^3$. Thus, the slugging frequencies observed in the present study are highly likely to be a function of the distributor design used (1x1 mm holes with a 26 μm aperture mesh). A future research direction could be to use additive manufacturing to optimize a distributor design to suppress unwanted flow regimes.

$$f = \frac{1}{\pi} \sqrt{\frac{g}{H_{mf}}}$$

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Figure 14 – Examples of the progression of the frequency spectra of the pressure drop signal when increasing the gas superficial velocity | stainless steel mesh distributor | (a) 5x5 mm, (b) 10x10 mm, (c) 15x15 mm | 93 ± 10 μm silica particles | $H/D_h = 2$ | frequency spectra maps for all particle and fluidized bed configurations are included in the supplementary materials document.

In the turbulent regime in the 5x5 mm bed, the dominant frequency can be seen to be increasing again. Note, the frequency spectra in Figure 14 were normalised between 0 and 1 for each gas flow rate. Therefore, although the dominant frequencies increased following the onset of fluidization, the intensity of this peak relative to the slugging regime also decreased. The increasing frequency and decreasing intensity are a consequence of the gas slugs breaking down into smaller bubble structures, approaching the steady release of the gas phase from the bed.
3.5 Flow Regime Maps

3.5.1 High Speed Camera Data

Figure 15a shows the flow regime transition boundaries observed qualitatively from the high speed camera data recorded in the six fluidized bed designs containing the silica particles using $D_h/D_p$ ratios of 2, 3 and 4. Here, the superficial gas velocities where a regime transition occurred were plotted against the ratio of bed hydraulic diameter to particle diameter $(D_h/D_p)$. Figure 15b also shows the result of smoothing these transition boundaries by plotting basic trend lines through the data to reveal a simple flow regime map for the additively manufactured fluidized beds. For reference, representative examples of each of the flow regimes observed are included in Figure 5 (animated .gif images are also available in the supplementary materials).

![Figure 15](image)

*Figure 15 – (a) Flow regime transition boundaries derived qualitatively from high speed camera videos, and (b) corresponding smoothed flow regime map | 93 ± 10 μm silica particles (Geldart A)*

It can be seen in Figure 15 that the boundary between particulate fluidization and bubbling (black line) asymptotically approached the minimum fluidization boundary (red line) as the ratio of $D_h/D_p$ increased. This means that wall effects become less apparent for larger bed diameters or small particle diameters, producing a faster emergence of the more desirable bubbling regime following minimum fluidization as the gas velocity is increased. Similar to Figure 10a, the minimum fluidization velocity (red line) decreased as the ratio of $D_h/D_p$ increased. This is again explained by the reduced wall effect, where friction is enhanced in the smaller beds because of the increased surface area to bed volume ratio. The superficial gas velocity required for slugging (green line) and turbulence (blue line) both increased with increasing $D_h/D_p$ ratio.

If the flow regime boundaries shown in Figure 15 are extrapolated to smaller ratios of $D_h/D_p$, it can be inferred that there will be a critical point around $D_h/D_p = 10$ where the minimum fluidization, slugging and turbulence velocities will merge. Here, turbulence will be produced immediately following the minimum fluidization point. Therefore, below this diameter ratio, fluidization would not likely be possible because the gas velocity to overcome wall adhesion would likely exceed the terminal velocity. This means for the 93 μm silica particles used in Figure 15, the smallest 3D printed channel that could observe fluidization would need a hydraulic diameter of at least $D_h = 0.93$ mm. Interestingly, this matches unpublished results from preliminary fluidization tests in our lab where clumping was observed in a 1 mm diameter 3D-printed fluidized bed containing 106–125 μm glass beads ($D_h/D_p = 8–10$).

3.5.2 Pressure Drop Data

Figure 16 plots the flow regime maps derived quantitatively using the pressure drop data using two sets of Geldart A particle (column 1) and two sets of Geldart B particle (column 2) at three particle bed
heights: $H/D_h = 2, 3$ and $4$ (row 1, 2 and 3 respectively). As with the data in Figure 15, basic trendlines (a range of power-law, polynomial and exponential lines of best fit) have been fitted to the data to create smoother flow regime maps, which are valid within the range of conditions considered. Note, in Figure 16a the results for the 10x10 mm and 15x15 mm bed designs using both the stainless steel mesh and porous plate distributor are plotted. This shows that although particles were partially supported by the distributor plate when using the stainless steel mesh in the larger bed designs, the flow regime transition points were not affected, and that packed bed, bubbling, transitional and slugging behaviours were still observable and that these flow maps are valid.
The flow regime map in Figure 15b was derived qualitatively using just the 93 μm silica particles, whilst Figure 16a was derived quantitatively using both the 82 μm glass beads and 93 μm silica particles. Nevertheless, it can be seen that the minimum fluidization and turbulence onset curves were similar, validating the reliability of both methods (visualisation and pressure drop); the slight differences between the two \( U_{mf} \) curves are attributed to the low resolution of the gas flow rates used to collect the high-speed camera images; the value of \( U_{mf} \) is interpolated using the pressure drop method. The main differences between Figure 15b and Figure 16a are the slugging onset curves, and differences in bubbling behaviour. Using the high-speed camera data, separate particulate and bubbling regimes were distinguished. Particulate fluidization corresponded to minimal bulk motion with non-uniform bubble generation at the bed walls, whilst bubbling involved uniform bubbling across the full channel cross-section with full motion of the bulk. However, this difference did not manifest within the pressure drop data, suggesting that there would not be a significant difference in kinetics when adjusting the gas velocity across this ‘boundary’. Further, in the pressure drop data, at \( D_b/D_f \geq 100 \), a transitional regime was identified from the pressure drop data that was attributed to the growth of larger bubbles prior to the onset of slugging, which did not seem to correspond to a physical change in the high-speed camera data. As mentioned above, this was observed using both the stainless steel mesh and porous plate distributors. It is possible that machine learning applied to the high-speed camera data might be more sensitive to this subtle regime change. However, for a fully-enclosed additively manufactured fluidized bed, the unpolished walls would only be translucent, meaning high-speed camera imaging would be less robust. Resultantly, the pressure drop method is the recommended approach because it provides statistical information about fluidization across the full bed.

Column 1 in Figure 16 shows the three flow regime maps derived for the Geldart A particles with static bed height ratios of \( H/D_b = 2, 3 \) & 4. At all height ratios, the minimum fluidization and minimum bubbling curves coincided, indicating no particulate fluidization occurred. Additionally, the turbulence boundaries were similar at all height ratios, matching the results of the high-speed camera data (Figure 15a). The minimum fluidization velocity increased slightly at smaller \( D_b/D_f \) ratios (< 75) whereas it remained relatively unchanged for larger \( D_b/D_f \) ratios (> 75), as already discussed in Section 3.3. With \( H/D_b = 2 \), the slugging onset boundary decreased with increasing \( D_b/D_f \) ratios, whereas for \( H/D_b \geq 3 \), the slugging boundary initially followed the minimum fluidization boundary until \( D_b/D_f = 50 \), before remaining constant with increasing \( D_b/D_f \) ratios. This shows that bubbling is not possible under the following conditions: \( H/D_b \geq 3 \) and \( D_b/D_f \leq 50 \); under these conditions there is more time for gas slugs to develop. The transitional regime identified in the frequency spectra maps were only observed in the larger fluidized beds, starting at \( D_b/D_f \geq 90 \) with \( H/D_b = 2 \), and \( D_b/D_f \geq 75 \) with \( H/D_b \geq 3 \), thus, the appearance of this transitional slugging regime corresponds with the transition between micro- and macro-scale fluidization behaviours discussed in Section 3.3. This transition zone is attributed to the growth of gas slugs with diameters around 0.66\( D_b-D_b \), matching the slugging definition of Kong et al. (2017), where wall friction starts to increase as the bubble size grows. The dominant frequency in the pressure fluctuations finally stabilise once full slug development has been reached (with \( D_{slug} \sim D_b \)).

The flow regime maps derived for the Geldart B particles are shown in column 2 in Figure 16. Unlike the Geldart A particles, the minimum bubbling velocities occurred before the minimum fluidization point at small \( D_b/D_f \) ratios (< 30), which was more prevalent at higher \( H/D_b \) ratios (> 3). This means gas bubbles were produced before the entire weight of the bed was supported by the gas; here, the solids likely ‘flow’ around the outer edges of the bubbles before re-settling after the transit of the bubble is completed. This could occur because there are larger void spaces between Geldart B particles, allowing greater movement than Geldart A particles without fluidization. The minimum fluidization velocities with the Geldart B particles were higher than the Geldart A particles in the smaller beds, but approached the Geldart A values with \( D_b/D_f = 100 \). This is because these particles were larger but had a similar density to the Geldart A particles, meaning they had a greater individual weight requiring greater drag
force from the gas to fluidize. Compared to the Geldart A particles, there was also a smaller operating window for bubbling, and the slugging onset point was lower at higher Dₚ/Dₚ ratios. The slugging onset velocity of the Geldart B particles might be lower because the Geldart A particles can pack closer together, resulting in a larger bubble break-up rate that counteracts bubble coalescence. The onset of turbulence also occurred at a lower gas velocity with the Geldart B particles. Turbulence signifies a loss of the distinction between the gas and solid phases. The larger weights of the individual Geldart B particles might reduce the particle-particle adhesive forces, reducing the analogous surface tension around the gas bubbles, resulting in a faster loss of bubble structures.

Another difference with the Geldart B particles with H/Dₕ ≥ 3 was the turbulent onset velocities seemingly decreased with increasing Dₚ/Dₚ, whilst the slugging velocities monotonically decreased before reaching a plateau. One physical explanation for the lower slugging boundaries, as mentioned above, is the Geldart B particles could stabilise/promote bubble coalescence because of the inherently larger gaps between the particles. For the decreasing turbulence onset point, this could just be an artefact of the limited sensitivity of the pressure transducer used for the experiments (± 500 Pa). I.e. it was not possible to perform fluidization experiments in the larger beds (Dₚ ≥ 10 mm) with particle heights of H/Dₕ ≥ 3 at the gas velocities needed to observe turbulence without also saturating the pressure transducer signal. Thus, the turbulence boundaries for H/Dₕ ≥ 3 are likely subject to increased uncertainty; this is accounted for by the addition of a dotted line that shows the turbulence boundary observed with H/Dₕ = 2.

Wang & Fan (2011) also proposed a flow map for Plexiglas fluidized beds containing 53 μm FCC particles, which was derived from a combination of their experimental data (using 0.7–5 mm diameter beds) and existing correlations from the literature for larger diameter beds. For Dₗ < 15 mm, their flow map shows largely the same features as Figure 16a: a large fixed bed region, narrow bubbling window, slugging and turbulence. Because Wang & Fan (2011) were able to use higher gas velocities in their experiments, they also measured the onset of fast fluidization, whose boundary occurred with an additional ~60 mm/s gas velocity beyond the turbulent onset boundary. Other notable differences with Wang & Fan’s (2011) map are: their fixed bed window did not become independent of the bed diameter until around Dₗ ~ 100 mm, and their particulate fluidization was observed even when using larger bed diameters (Dₕ > 100 mm).

4 Conclusions
Additive manufacturing (AM) presents a new opportunity to explore unique fluidized bed geometries that would have previously been difficult/impossible to construct using conventional manufacturing methods. Some potential examples include the fabrication of more complex bed cross-section shapes, adding baffles or other wall surface features to promote better fluid percolation and/or fluid-solid mixing, sub-dividing the freeboard region into different compartments to realise multi-stage configurations, and optimization of the distributor assembly. There is also the option to either 3D-print single bespoke fluidized beds (as in this study) or 3D-print modular fluidized bed components to realise plug-and-play setups for rapid and high-throughput screening applications. However, depending on the specific AM technology, the printed surfaces can exhibit greater roughness than conventional materials, necessitating the study of fluidization in these 3D printed devices.

In this study, for the first time, 3D printing has been used to successfully fabricate mini and micro fluidized beds. Four sizes of glass and silica particles exhibiting both Geldart A and Geldart B characteristics were subsequently fluidized in six fluidized bed designs (possessing hydraulic diameters, Dₚ, ranging from 3 to 15 mm) to map the fluidization characteristics. Using pressure drop data, it was found that a bed diameter to particle diameter ratio of Dₚ/Dₕ = 75 denotes the transition point from micro-scale to macro-scale fluidization characteristics in these 3D-printed fluidized beds. This is significantly lower than the conventional PMMA fluidized beds reported in the literature, where the transition is closer to Dₚ/Dₕ = 300, showing that additive manufacturing has a useful future role for translating lab scale data to larger scales with lower waste generation (i.e. lower material usage).
Additionally, a new criteria for slugging has been proposed using only pressure drop data. The method involves creating normalised 2D maps of the frequency spectra as the gas velocity is increased. The onset of slugging is taken as the point where the average dominant frequency becomes stable. Using this method, a transitional regime between bubbling and slugging could also be identified for the Geldart A particles. Using the pressure drop data, a comprehensive set of flow regime maps were derived that should enable researchers to configure 3D printed fluidized beds for a desired flow regime for a particular process (where the gas velocity can be selected based on the residence time).

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Nomenclature

- \( A \): Cross-sectional area of the particle bed perpendicular to the gas flow (m²)
- \( D_h \): Bed hydraulic diameter for non-circular cross-sections (= \( 4A/P \)) (m)
- \( D_p \): Particle diameter (m)
- \( D_t \): Bed diameter for circular cross-sections (m)
- \( f \): Bubbling/slugging frequency (Hz)
- \( H_{mf} \): Height of the particle bed at minimum fluidization (m)
- \( H_s \): Static bed height (m)
- \( g \): Gravitational acceleration (9.81 m.s⁻²)
- \( k_1 \): Empirical constant
- \( k_2 \): Empirical constant
- \( m \): Total mass of the particle bed (kg)
- \( P \): Perimeter of the fluidized bed (m)
- \( U_c \): Critical velocity for onset of turbulence (m.s⁻¹)
- \( U_g \): Gas velocity (m.s⁻¹)
- \( U_{mf} \): Minimum fluidization velocity (m.s⁻¹)
- \( V_p \): Total volume of the particles (m³)
- \( V_{t,mf} \): Total volume of the bed (including voids) at minimum fluidization (m³)

Dimensionless Groups

- \( \text{Ar} \): Archimedes Number (= \( \phi D_p \frac{3}{4} (\rho_p - \rho_g) g \rho_g / \mu_g^2 \))
- \( \text{Re} \): Reynolds Number (= \( \rho_g (\phi D_p) U_{mf} / \mu_g \))

Greek Letters

- \( \Delta P_b \): Pressure drop due to buoyant weight of the particle bed (Pa)
- \( \Delta P_{\text{Ergun}} \): Pressure drop predicted by the Ergun equation in the packed bed regime (Pa)
- \( \varepsilon \): Voidage
- \( \varepsilon_{mf} \): Voidage at minimum fluidization
- \( \mu_g \): Gas viscosity (Pa.s)
- \( \rho_g \): Gas density (kg.m⁻³)
- \( \rho_p \): Particle density (kg.m⁻³)
- \( \phi \): Particle friction angle
- \( \phi \): Particle sphericity

References


S. Laín, M. Sommerfeld. Euler/Lagrange computations of pneumatic conveying in a horizontal channel with different wall roughness. Powder Technology 184 (2008) 76-88


P.N. Loezos, P. Costamagna, S. Sundaresan. The role of contact stresses and wall friction on fluidization. Chemical Engineering Science 57 (2002) 5123-5141


