SOLID FLUX MEASUREMENT IN A SOLID-LIQUID MICRO-CIRCULATING FLUIDISED BED

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Abstract – Solid-liquid circulating fluidised beds (CFB) have found industrial applications in many processes due to their good characteristics such is excellent particle-liquid contact, but have not yet been applied in the micro technology context. Solid-liquid micro circulating fluidised beds (µCFBs), which essentially involve fluidisation of micro-particles in sub-centimetre beds, hold promise of applications in the microfluidics and micro-process technology context. From an engineering standpoint, it is vital to know the solid flux, since that dictates the bed capability and operability as processing equipment. Albeit there are several studies on solid flux measurement in CFBs, we are presenting the first experimental study on solid flux measurement in a CFB at micro-scale. The experimental research was performed in a micro-CFB which was made by micro-machining channels of $1mm^2$ cross section in Perspex. PMMA and soda lime glass micro-particles were used as the fluidised particles and tap water as the fluidising liquid. The digital particle image velocimetry (PIV) method was used as a novel measurement technique to measure the solid flux in the micro-CFB system. The critical transition velocity, U_{cr}, is approximately equal to the particle terminal velocity, U_t. As in macroscopic CFBs system, U_{cr} decreased with solid inventory (1-9%) before levelling off at a high enough solid inventory (10-25%) and it increases with reduction in particle size and density

INTRODUCTION

Solid-liquid circulating fluidised beds are an important solid-liquid processing technology in various industrial processes due to their advantages such as improved heat and mass transfer, reduced back mixing, good liquid-solid contact, and good control of reaction and regeneration of catalyst or bio-solid at the same time (Gnanasundaram et al., 2014, Vidyasagar et al., 2011). They have found applications in biochemical, hydrometallurgy, environmental, wastewater treatment, mineral processing and petrochemical industries as reactors, bio-reactors, crystallizers and heat exchangers (Liang et al., 1995, Natarajan et al., 2014). In spite of this, circulating fluidised beds have not yet been applied in the micro technology context. On other hand there are several studies of a micro-liquid-fluidized beds mainly concerning the hydrodynamics.

Micro-fluidised beds refers to fluidised bed with the bed cross-section or inner diameter at micrometre scale (Potic et al., 2005, Zivkovic et al., 2013, do Nascimento et al., 2016) with potential applications in a micro technology and micro fluidics context (Kivikero et al., 2011, Yu et al., 2013, Geng et al., 2013, Yang et al., 2016, Pereiro et al., 2017). These types of bed are considered to be a promising way of achieving high quality fluid and solid mixing and process intensification of heat and mass transfer under a laminar flow regime and could potentially offer novel process opportunities (Doroodchi et al., 2012, Hessel et al., 2014). Micro-reactors in general are considered to be ideal for performing reactions in circumstances which would normally be limited by heat and mass transfer and unsafe operations (Reay et al., 2013). Micro-fluidised beds combine the advantages of fluidised beds and micro technology systems such as reduced pollution, waste and by products, less energy and resources consumption, less operational and capital cost, increased safety, intensive heat and mass transfer, and increased chemical reaction conversion rates, as well as exhibiting good mixing and temperature uniformity. All these important qualities make micro-fluidised beds more efficient and sustainable fluid-solid processing equipment. In addition, in some applications continuous circulation of particles

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would be required such as continuous regeneration of catalyst or bio-solid particles for which circulating fluidised beds are excellent technology (Chavan et al., 2009).

Albeit there are few reports on liquid-solid micro-fluidised beds as outlined above, the paper presents the first experimental study of a solid-liquid circulating fluidised bed at the microscale. It is well know that particle handling in micro technology devices remains one of the big challenges in the field (Jensen, 2001). Development of a micro circulating fluidised bed is providing one solution to the problem, e.g. for solid catalyst recovery, recycle and regeneration (Golriz et al., 2003, Chavan et al., 2009). Measurement of solid flux in a micro circulating fluidised bed are extremely hard to be implemented using current measurement technology such as valve, X-ray tomography, electrical capacitance tomography, , magnetic resonance imaging, as they are expensive, but more importantly it is very difficult to scale down for application in a microfluidics context (Wang and Fan, 2011). In order to successfully design a solid-liquid micro circulating fluidised bed system for micro-technology applications, it is essential to understand their hydrodynamics such as solid flux as it determines the bed performance as processing equipment, controlling heat and mass transfer, and dictating mixing in the system (Dietrich et al., 2013).

The solid circulation rate measurement in a solid-liquid circulating fluidised bed has been reported previously by various researchers. Several research groups (Liang et al., 1997, Zheng et al., 1999, Vidyasagar et al., 2008, Natarajan et al., 2008) measured the solid circulation rate by using a simple procedure with valve in downcomer. The accumulation of particles above the ball valve at given time interval could be measured by closing valve in downcomer, thus giving the particle circulation rate. They reported that the solid circulation rate increases with liquid flow rate and solid feed pipe diameter but decreases with increase particle density and size. Roy *et al.* (2001) used a radioactive technique to measure the solid circulation rate. The basic theory was to determine the particles volumetric flow rate by measuring the particles velocity and volume fraction in the downcomer. Particle volume fraction was obtained by densitometry measurement, while particle velocity was obtained by measuring the falling time of a single radio-active tracer particle between two detectors installed in the downcomer. Wang *et al.* (1998) used electrical capacitance tomography to measure the solid circulation rate in a circulating fluidised bed while Hensler *et al.* (2016) employed X-ray tomography to determine the solid circulation rate.. Wu *et al.* (2001) developed and employed an impact plate flow meter to determine the solid circulation rate by measuring the torque on a hinged plate from falling particles impacts.

In the current research investigation particle imaging velocimetry (PIV) technique using open-source PIVlab software (Thielicke and Stamhuis, 2014) was used to determine the solid flux in the micro-circulating fluidised bed. Digital PIV technique has been employed previously by researchers for measuring the flow velocity profiles in granular flow (Sarno et al., 2014, Tebianian et al., 2015). The advantage of the digital PIV technique over other conventional techniques such as the valve technique is that the technique is non-invasive and easy to implement in microfluidics setup which is not trivial for the valve. The effects of operating parameters such as solid inventory, particle size and density on solid flux has been carefully studied using PIVlab and MatLab software.

EXPERIMENTAL DETAILS

The schematic representation of the research experimental set up is illustrated in Fig. 1(a). The system consists of a syringe pump (AL-4000, WPI INC., US) to pump the water as a fluidising medium at the desired flow rate using a 5ml B-D Plastipak syringe, and Euromex Nexius trinocular digital microscope fitted with a USB digital camera (JB Microscopes Ltd, UK) to record the micro fluidisation behaviour. The images and movies were saved on a computer for offline studies. The system used in the present experimental investigation was made by micro machining channels in Perspex as schematically shown in Fig. 1(b). The micro-circulating fluidised bed consist of a riser column of 1 mm square cross-section and 100 mm in height, a solid-liquid separator, a downcomer acting as a particle reservoir, a solid return pipe, and a solid feeding pipe. At the base of the riser is the distributor (a 1.5 mm thick porous plate

distributor with mean pore size of 21 μm) which prevents particles leaving the bed at the bottom and provides uniform flow distribution and stable fluidisation. The solid-liquid separator is a simple diamond shaped expansion that enables the particles to be separated from the outflowing liquid.

Two different groups of particles were used as fluidised solid: (1) soda lime glass microspheres of five different diameters, d = 26 ± 1.5, 30 ± 1.5, 35 ± 3, 58 ± 5, and 115 ± 9 μm whose density is ρ_p = 2500 kg/m³ and (2) PMMA particles of five different diameters, d = 23 ± 3.5, 35 ± 3, 41 ± 3.5, 58 ± 5, 115 ± 9 whose density is ρ_p = 1200kg/m³. Tap water (density ρ_f=998 kg/m³ and viscosity μ_f= 0.003 Pa.s) was used as the fluidising liquid. All experiments were performed at room temperature of average 18 ± 2 ºC. The Stokes particle terminal velocity for laminar flow which is applicable to the research investigation as Re<1 is given by

$$U_t = \frac{(\rho_p - \rho_f)gd_p^2}{18\mu} \qquad (1)$$
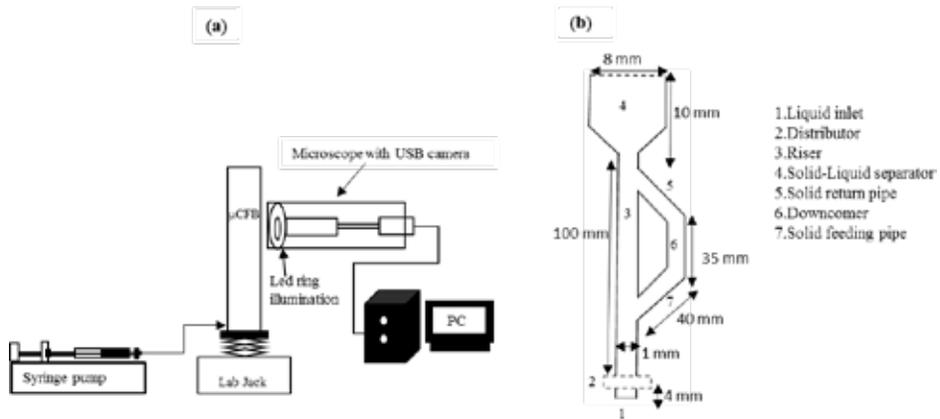
where g is gravitational acceleration.

Fig. 1. Schematic of (a) experimental set-up for imaging and (b) micro-circulating fluidised bed.

EXPERIMENTAL METHODOLOGY

Initially the bed was filled with liquid (water) using a syringe pump and the bed was packed with particles. The solid inventory was measured with the aid of ImageJ (Schneider et al., 2012) in terms of surface percentage occupied by the particles out of the whole system surface (in this case this is the same as volume percentage as the depth is constant). Liquid at varying velocity was pumped by a syringe pump from the syringe to the bed inlet to produce the fluidisation liquid at the required superficial velocity in the bed. When the liquid flow rate was high enough, particles were carried out of the riser and separated from the outflowing liquid by the solid-liquid separator and recirculated back to the riser through the solid feed pipe. The experimental procedure was performed with both decreasing and increasing superficial liquid velocities. For each particle, this procedure was repeated at least three times to ensure repeatability and the measurement of experimental errors. Digital movies of liquid-solid fluidisation behaviour were recorded and stored on a PC for off-line analysis. The off-line examination comprised of movies conversion into successive frame sequences by VLC media player, and particle displacement calculation from successive frames using PIVlab, a Matlab code, which determines the velocity of particles by cross correlation PIV algorithm of multiple small sub-images.

Determination of particle displacement using the PIVlab code in the current research investigation is summarised as follow. The downcomer was chosen as the region of interest and a mask was applied to exclude the outside regions from the analysis as shown in Fig. 2. Image pre-processing contrast limited adaptive histogram equalisation (CLAHE) was applied to all frames to increase the image contrast and the probability of detecting valid vectors. Each image was divided into small sub images (interrogation region), and cross-correlation of successive frames was applied to determine the most likely

displacement of particles in these region. Particle displacement was determined using FFT (fast Fourier transform correlation with multiple pass and deforming windows algorithm). A three step cross correlation analysis with an interrogation area of 64 pixel in first pass, 32 pixel in second pass, and 16 pixel in the third pass (corresponding to 0.64, 0.32, and 0.16 mm in real scale respectively) was chosen to determine the particle displacement of the image data, and each interrogation area was overlapped by 50%. The displacement information obtained in each pass is used to shift the interrogation windows in the next pass to increase the resolution of vector map, signal to noise ratio without sacrificing robustness. Image calibration was performed by specifying the bed cross sectional area (1 mm) and the time step between images of one frame (25 frames per second). The vector field were smoothed and validated using a number of filters (local median filter, and standard deviation filter) while missing data was interpolated using the boundary value solver interpolation technique to increase the accuracy of the velocity calculation. All the above procedure was repeated to calculate the velocity (horizontal u and vertical v component) for each frame for all the PMMA and glass particle at various liquid flowrate. The results obtained using PIVlab code was exported as a consecutive Mat-file into Matlab workspace for further processing. Matlab was then employed for adjusting velocity field due to background movement and for time averaging of velocity field for each particle at different liquid flow rate by averaging the instantaneous velocity obtained using PIVlab code.
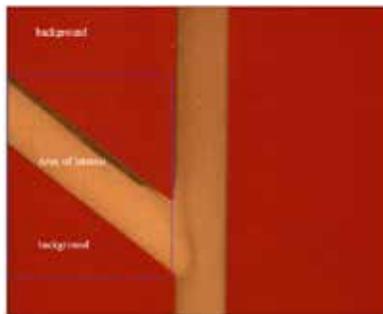

Fig. 2. Typical image of micro-circulating fluidized bed for the 58 µm glass particles at 445 µl/min flow rate inside 15% solid inventory bed showing areas of interest for the digital PIV analysis.

RESULTS AND DISCUSSION

Background movement

Initial PIVlab image analysis shows that particles moves upwards as shown in Fig. 3(a) instead of downwards along the solid return pipe as expected and visually observed. Further analysis showed considerable background movement so it was necessary to calculate the background movement velocity field using PIVlab as shown in Fig 3(b). The imported vector field of particles in downcomer in Matlab is shown in Fig. 4(a) with the erroneous upward motion of particles. The velocities were then correctly adjusted by subtracting the background motion (Fig. 3(b)) in Matlab to obtain the correct particle velocity field with downward motion as shown in Fig. 4(b).
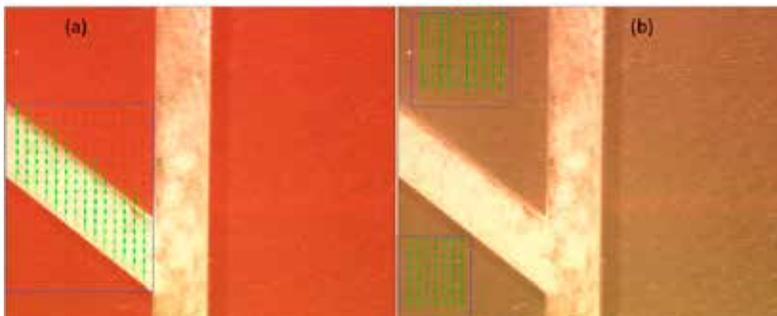

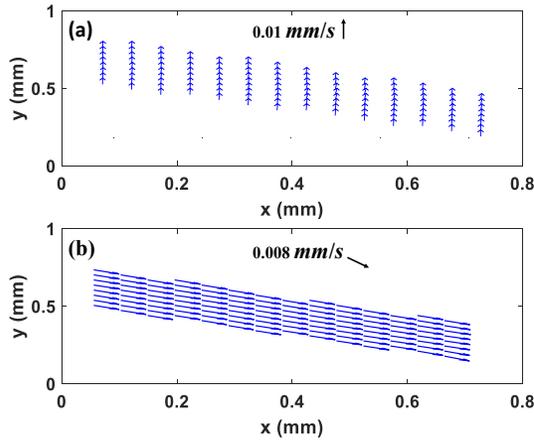Fig. 3. Velocity vector field in (a) the downcomer and (b) in background determined by PIVlab analysis.

Fig.4. (a) Original particle velocity field and (b) after subtraction of the background movement for the 58 µm glass particles at 445 µl/min flow rate inside 15% solid inventory bed.

Effect of the liquid flow rate on solid flux

In solid-liquid circulating fluidised beds, particle motion is controlled by changing the inlet superficial liquid flow rate. Fig. 5 displays the effect of liquid velocity on the solid flux for glass and PMMA particles as determined by digital PIV analysis described in previous section. The experimental results indicate that the solid flux in the system is close to zero (no solid movement) at low enough velocities then increases sharply at some critical superficial liquid velocity and finally plateaus at higher superficial liquid flow rate. The change in solid flux with superficial liquid flow rate indicates two distinct zones. The first zone (initial circulating fluidisation zone) where solid flux increases rapidly with increasing superficial liquid flow rate and the second zone (fully developed zone) where solid flux insignificantly varies with increasing superficial liquid velocity as reported by Zheng *et al.* (1999). The critical transition velocity from conventional fluidised bed to circulating fluidised beds occurs at the point where the solid circulation rate becomes zero with reducing superficial liquid velocity. Thus, the critical transition velocities are determined as the intercept of no particle flow (nearly zero) and initial circulating zone line as shown in the plots. These plots clearly also show that the critical transition velocity, U_{cr}, is approximately equal to the particle terminal velocity, U_t in all consider cases.
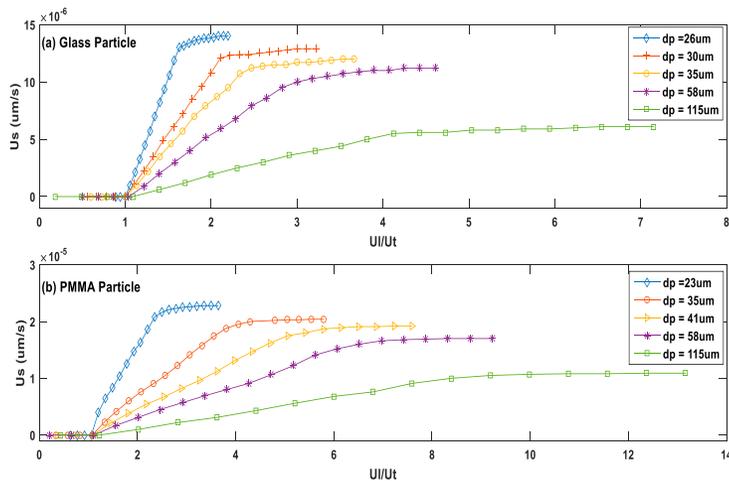

Fig.5. Particle circulating speed as a function of normalised velocity (U_l/U_t) for (a) glass and (b) PMMA particle. Error are smaller than the symbols.

Effect of solid inventory in the system

Fig.6 shows the normalised critical transition velocity as a function of solid inventory, indicating that in the solid-liquid micro circulating fluidised bed the critical transition velocity is strongly influenced by solid inventory. For beds with a solid inventory lower than 10 %, the transition from conventional to circulating fluidised bed is much larger velocities than the particle terminal velocity (U_{cr}/U_t = 1.5 to 2.5). However, for systems with a solid inventory higher than 10%, the critical transition velocity from conventional to circulating fluidised bed regime occurs close to the particle terminal velocity, and the normalised transition velocity is approximately 1. These observations are similar to those reported by Liang *et al.* (1997), the critical transition velocity decreases with solid inventory and finally becomes stable when the solid inventory is high enough. On other hand, our findings are not in line with results of Zheng and Zhu as their reported onset velocity (U_{cf}) which gives the lowest critical transition velocity from the conventional to circulating fluidised bed regime was found to be independent of the bed geometry, operating conditions and solid inventory probably due to the method applied.
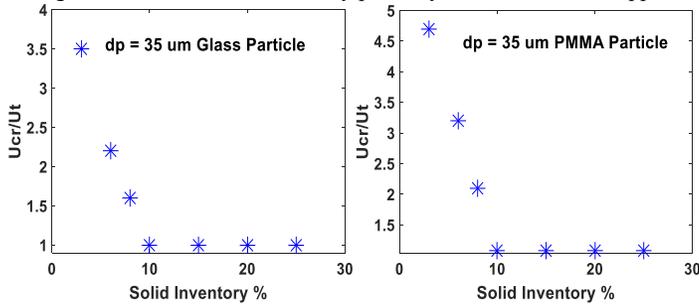

Fig. 6. Effect of solid inventory on normalised transition velocity for 35μm (a) glass and (b) PMMA particles.
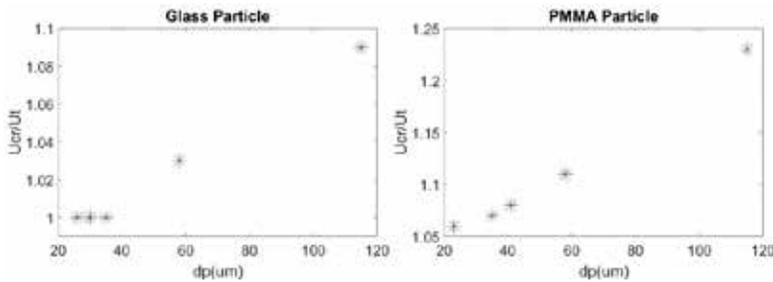

Fig. 7. Normalised critical transition velocity as a function of particle size for (a) glass and (b) PMMA micro-particles in beds of high solid inventory (in the range of 10 - 25%).

Effect of particle size

Fig. 7 shows that the transition from conventional to circulating fluidised bed regime in a solid-liquid micro circulating fluidised bed is influenced by particle properties such as size and surface properties beyond the influence on the particle terminal velocity. First there is a trend of increased normalised critical transition velocity with an increase in particle size which is probably because of increased wall effects which are not usually present in large circulating fluidised beds. It can also be observed that the normalised transition velocity (U_{cr}/U_t) is considerable higher for PMMA particles compared to glass particles of the same size. This is probably due to difference in surface properties with PMMA particles being hydrophobic while glass particles hydrophilic. Particle agglomeration due to cohesion was visually observed for PMMA and some particle adhesion to the walls. Cohesion increases particle size as a result of agglomeration and that may postpone the critical transition velocity (U_{cr}) from conventional to circulating fluidised bed regime and the wall adhesion present in the downcomer will also contribute to postponing of the transition.

CONCLUSION

The digital PIV analysis using PIVlab and MATLAB software was used to determine the solid flux in a micro-circulating fluidised bed. The implemented digital PIV technique to estimate the solid flux seems promising, and the results looks relevant when comparing with previous reported studies. As in macroscopic circulating fluidised bed, the solid flux in micro-circulating fluidised bed increases with liquid velocity in two distinct zone increasing sharply first then levelling off at higher inlet fluid velocities. The determined transition velocities from solid flux versus velocity plots are comparable to the particle terminal velocity, i.e. the normalised transition velocity is approximately 1 in line with previous studies. The transition velocity from is strongly influenced by solid inventory, i.e. it decreases with solid inventory before levelling off at high enough solid inventory. Finally, it was observed a weak increase in the normalized transition velocity with particle size which is probably due the wall effects (higher particle to bed ratio).

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