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EXPERIMENTAL STUDY OF A LIQUID FLUIDIZATION IN A MICROFLUIDIC CHANNEL

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Introduction

Microfluidics¹⁻³ is the science and technology of processing and manipulation of small amounts of fluids in conduits having dimensions of the order of tens to hundreds of micrometers. Just as integrated microelectronic circuits revolutionized computation by enormously reducing the space, labor, and time required for complex calculations, microfluidics holds promise in the automation of chemical and biochemical analyses and processing as well as in the field of medical diagnostics⁴⁻⁶. However, transport processes in microfluidic systems are dominated by molecular diffusion as the flows are almost universally laminar. This has the potential to over-shadow many of the benefits of miniaturization for applications such as micro-reactors,^{7,8} which is one of the areas in chemical engineering where microfluidics is said to offer great potential.^{9,10} Fluidized beds have long been used at the macroscale to substantially enhance mixing, heat and mass transfer. Recent simulation work has shown that they may also offer similar benefits at the microscale.^{11,12} In addition, the use of small microparticles would offer significant surface area per unit volume, making microfluidized beds ideal in diagnostics and similar contexts.^{13,14}

The concept of a liquid micro-fluidized bed was introduced recently to refer to beds with inner diameters of a few millimetres¹⁵, though the 'micro' prefix in this case is used in the traditional sense of a small tubular reactor in the reactor engineering context. However, nowadays with the widening use of micro-fabrication techniques this prefix generally refers to fluidic systems on micro-chips with micro-channels of non-circular geometry, usually rectangular cross-section⁷. Furthermore, surface forces are more important for channels smaller than 1 mm, which may be considered as a coarse boundary between micro- and macro-scale flows.^{16,17} The already-mentioned experimental study¹⁵ and more recent work¹⁸ were performed in capillaries of around 1 millimetre in

size (1 mm and 0.8 mm respectively) which puts them on the boundary between macro- and micro-scales. Here we report the experimental study of liquid fluidization of approximately 30 μm glass microparticles in a rectangular microfluidic channel with a cross-section of 400 μm x 175 μm – as far as we are aware, this is the first such experimental realisation of a fluidized bed in channels typical of microfluidics. The aim of the study reported here was to first realise such a microfluidized bed and then examine the validity of the classical fluidized bed design equations for it. This was achieved by determining minimum fluidization velocity (U_{mf}) and superficial velocity-voidage relationship experimentally and comparing them with predictions of models that are typically used at the macroscale.

Experimental details

Microfluidized bed

The microfluidized bed used here is shown in Fig. 1. The micro-channel from which the bed was formed was fabricated in a polydimethylsiloxane (PDMS) ‘chip’ by standard soft lithography techniques.¹⁹ The width of the channel was 400 μm , and depth 175 μm . The distributor of the fluidized bed, which was designed using computational fluid dynamics,^{20,21} consisted of eight 40 μm wide pillars separated by 9 μm gaps.

Fig. 1. Optical micrograph showing the microfluidized bed with the distributor composed of eight pillars of 40 μm width separated by 9 μm gaps.

The chip containing the microfluidized bed was mounted vertically in a custom made holder on a height adjustable rotary stage (M488, Newport Corporation, US) for easy manipulation, Fig. 2. A trinocular stereomicroscope with holder and fibre optic illuminator (SMZ-168-TH, Motic, China) connected to a digital camera (KY-F550E, JVC, Japan) was used for recording images. The images were stored on a PC for offline

analysis. The fluidizing liquid was pumped by the PHD ULTRA syringe pump (Harvard Apparatus, US) at desired flow rates. Experiments were performed at an ambient temperature of 25°C.

Fig. 2. Schematic of experimental setup for the top-view flow visualization.

Fluidizing material

The bed was composed of as-supplied soda lime glass microspheres of diameter $d = 30.5 \pm 1.5 \mu\text{m}$ and density $\rho = 2500 \text{ kg/m}^3$ (Cospheric LLC, US). Both deionized water, surfactant-containing deionized water and ethanol were used to fluidize the particles. The water was obtained from an ELGA PURELAB (VWS Ltd., UK) classic water purification system. The anhydrous ethanol ($\geq 99.5\%$) and the non-ionic surfactant Tween 80 used was as-supplied by Sigma Aldrich (St. Louis, Missouri, US).

Results

Water as a fluidizing medium: adhesion problem

Initial efforts to fluidize the glass particles in the microfluidized bed using deionized water proved impossible due to the particles adhering to the PDMS walls of the micro-fluidized bed as shown in Fig. 3. Subsequently, a non-ionic surfactant (Tween 80) was used in an attempt to minimize the effect of adhesive forces relative to the buoyancy and gravity forces.²² However, adhesion still occurred, albeit to a lesser extent, with the micro-particles only being observed to flow at superficial velocities several times the particle settling velocity (approximately $750 \mu\text{m/s}$), leading to their rapid elution from the bed. This indicates that whilst the surfactant lead to some decrease in the particle-particle and particle-wall adhesion forces as anticipated,²² it was still insufficient to enable fluidization.

Fig. 3. Optical micrograph showing adhesion of glass particles to PDMS walls of the micro-channel when deionized water is used as the fluidizing medium

Fig. 4. Optical micrographs of the micro-fluidized bed at different ethanol volumetric flow rates. Initial height $h_0 = 3.215 \pm 0.015$ mm (see video online for greater detail).

Ethanol as a fluidizing medium

Ethanol proved to be a good fluidizing medium for the glass particles used here in the PDMS micro-channel, with smooth, stable homogenous fluidization behaviour (see video in supplementary information. Fig. 4 shows photos of the bed expansions of the particles at different ethanol flow rates. There was a sharp flat interface between the top of the bed and freeboard as typical for liquid fluidized bed. Particle adhesion to the microchannel walls was not observed.

Expansion experiments were performed starting from the highest flow rate and then incrementally decreasing the flow rate to avoid pressure overshoot, which could be present due to the small channel-to-particle diameter

ratio (approximately 8 based on hydraulic diameter of micro-channel, $D_h = 243.5 \mu\text{m}$).¹⁸ At flow rates at or below 200 nl/min, minor particle leakage through the distributor was observed (see video); this was taken into account in the subsequent analysis. Two cycles of defluidization were performed; no further were possible with the chip due to failure of the pillars that constitute the distributor.

Fig. 5. Relative bed height $(h-h_0)/h_0$ as a function of ethanol superficial velocity, U , for two experimental runs: squares (1st run, $h_0 = 3.335 \text{ mm}$) and circles (2nd run, $h_0 = 3.215 \text{ mm}$).

ImageJ²³ was used for offline image analysis to obtain the height of the fluidized bed as a function of ethanol flow rate, which is shown in Fig. 5 for two defluidization experiments. The fact that the data from the two experiments are essentially identical indicates the fluidization behaviour is quite repeatable. The noticeable particle leakage at superficial velocities just above the incipient fluidization point meant the minimum fluidization velocity, U_{mf} , had to be determined by extrapolating Fig. 5 to the packed bed height. Although this introduces some uncertainties, the estimated minimum fluidisation velocity of $U_{mf} = 17.25 \pm 1.35 \mu\text{m/s}$ falls within the range of $18.3 \pm 3 \mu\text{m/s}$ predicted by the Ergun equation²⁴ using an estimated packed bed voidage²⁵ of $\epsilon_{mf} = 0.46 \pm 0.02$. This somewhat higher bed porosity than the value typically used for packed beds in fluidization experiments, $\epsilon_{mf} = 0.426$, is a consequence of the significant effect that the wall has on the packing here relative to typical fluidised beds where the channel-to-particle diameter ratios are much larger.

Knowledge of the bed expansion characteristic (*i.e.* the relationship between the liquid superficial velocity, U , and bed voidage, ε) is crucial as bed voidage significantly influences the mixing, heat and mass transfer characteristics of fluidized beds. Using the data in Fig. 5, the bed expansion characteristic for the micro-fluidized bed shown in Fig. 6 was estimated using

$$\varepsilon = 1 - (1 - \varepsilon_{mf}) \frac{h_0}{h} \quad (1)$$

where $\varepsilon_{mf} = 0.46 \pm 0.02$ ²⁵. The bed expansion data in this figure is well described by the well-known empirical Richardson-Zaki equation,²⁷ which can be expressed as

$$U = k U_t \varepsilon^n \quad (2)$$

where U_t is the particles settling velocity, k a parameter that was set to unity in the original work of Richardson and Zaki²⁷ that was concerned with the laminar regime, and n an exponent that takes a value of 4.65 in their original work. Here we obtain from the slope of the line in Fig. 6 an exponent value of $n = 5.07 \pm 0.30$, which is clearly greater than that of the original Richardson and Zaki²⁷ work, but closer to a value of 4.9 obtained by Fan *et al.*²⁸ for fluidization of glass microsphere (the smallest being 54 μm) in a macroscale bed.

Extrapolating the experimental data to a voidage of unity gives $1600 \pm 60 \mu\text{m/s}$, which is almost double the theoretical Stokes law terminal velocity, $U_t = 836 \mu\text{m/s}$. This suggests that for the micro-fluidized bed considered here $k = 1.91 \pm 0.07$, which is in stark contrast with macroscopic experiments where $k \leq 1$ in general²⁶, with only Fan *et al.*²⁸ obtaining k values slightly above unity. Although not mentioned by them, analysis of the experimental data of Potic *et al.*¹⁵ for water fluidization in an 800 μm diameter capillary gives values similar to that obtained by us here ($k = 1.70 \pm 0.35$). These higher than usual values of the empirical constant, k , are probably due to the greater significance of surface forces in microfluidics, although a small de-fluidized zone near the distributor may also play some part, at least in our experiments. Fan *et al.*²⁸ did observe a change of slope at high voidages not considered in our work here – if this does occur, a second k value will be obtained that is closer to unity. Further investigation of this and other points raised above is underway and will be reported in a fuller paper in due course.

Fig. 6. Micro-fluidized bed expansion characteristics of 30 μm glass particles inside a 400 x 175 μm microfluidics channel. Line is the best-fit to the Richardson-Zaki model, Eq. 2 with lower and upper prediction bounds (segmented lines). Bars represent error in experimental points (y-errors smaller than symbols).

Conclusion

Fluidization of approximately 30 μm particles within a micro-channel of 175 x 400 μm rectangular cross-section using ethanol as a fluidizing medium demonstrated for the first time as far as we are aware that fluidization is possible in what can be truly called micro-channels. Comparison of the behavior observed in this micro-fluidized bed system with the conventional equations for fluidization shows that these equations are a good starting point for the design of micro-fluidized beds. The Ergun equation,²⁴ is applicable for evaluating the minimum fluidizing velocity provided the effect of the small particle/micro-channel diameter on the packed bed voidage is accounted for. The original empirical Richardson and Zaki²⁷ correlation, equation (2), also describes the micro-fluidized bed behavior observed here well, although higher values of empirical constants (k and n) than observed in more traditional fluidization are obtained – this requires further elucidation. Finally, fluidization was impossible when using deionized water as a fluidizing medium, demonstrating the importance of considering surface forces in true micro-fluidized bed systems.

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