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Experimental Study of a Liquid Fluidization in a Microfluidic Channel

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Significance

This study reports the first ever experimental study of true micro-fluidization: a liquid-fluidized bed of 30.5 μm micro-particles within a rectangular micro-channel of 400 × 175 μm^2. The failure to fluidize the micro-particles with water demonstrated the importance of surface forces for micro-fluidization. Use of ethanol instead leads to fluidization that is largely well described by conventional fluidization equations, although higher values of empirical constant (k and n) are obtained for Richardson-Zaki correlation.

Keywords: fluidization, granular materials, liquid fluidized bed, microfluidics, microfluidized bed, multiphase flow

Introduction

Microfluidics[1–3] 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 diagnostics.[4–6] 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 microreactors,[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 microfluidized bed was introduced recently to refer to beds with inner diameters of a few millimetres,[15] although 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 microfabrication techniques, this prefix generally refers to fluidic systems on microchips with microchannels of noncircular geometry, usually rectangular cross-section.[7] Furthermore, surface forces are more important for channels smaller than 1 mm, which may be considered as a coarse boundary between microscale and macroscale flows.[16,17] The already-mentioned experimental study[15] and more recent work[18] were performed in capillaries of around 1 mm in size (1 and 0.8 mm, respectively) which puts them on the boundary between macroscale and microscale. Here, we report the experimental study of liquid fluidization of ~30-μm glass microparticles in a rectangular microfluidic channel with a cross-section of 400 × 175 μm^2—as far as we are aware, this is the first such experimental realization of a fluidized bed in channels typical of microfluidics. The aim of the study reported here was to first realize 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 microchannel 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,

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Additional Supporting Information may be found in the online version of this article.

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which was designed using computational fluid dynamics,[20,21] consisted of eight 40-μm wide pillars 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, USA) for easy manipulation, Figure 1. A trinocular stereomicroscope with holder and fiber 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, USA) at desired flow rates. Experiments were performed at an ambient temperature of 25°C.

Fluidizing material

The bed was composed of as-supplied soda lime glass microspheres of diameter $d = 30.5 \pm 1.5$ μm and density $\rho = 2500$ kg/m^3 (Cospheric LLC, USA). 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 nonionic surfactant Tween 80 were used as-supplied by Sigma Aldrich (St. Louis, MO).

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 microfluidized bed as shown in Figure 2a. Subsequently, a nonionic 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 microparticles only being observed to flow at superficial velocities several times the particle settling velocity (~750 μm/s), leading to their rapid elution from the bed. This indicates that while 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.

Ethanol as a fluidizing medium

Ethanol proved to be a good fluidizing medium for the glass particles used here in the PDMS microchannel, with smooth, stable homogenous fluidization behavior (see Supporting Information, Video S1). Figure 2b 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.

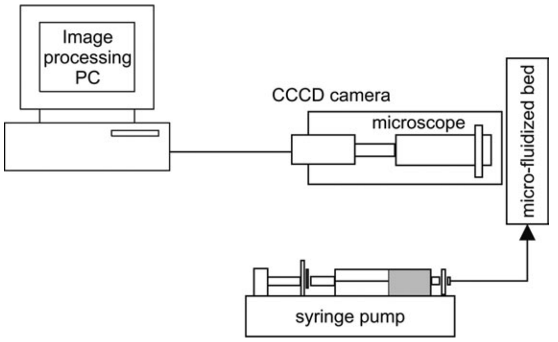

Figure 1. Schematic of experimental setup for the top-view flow visualization.

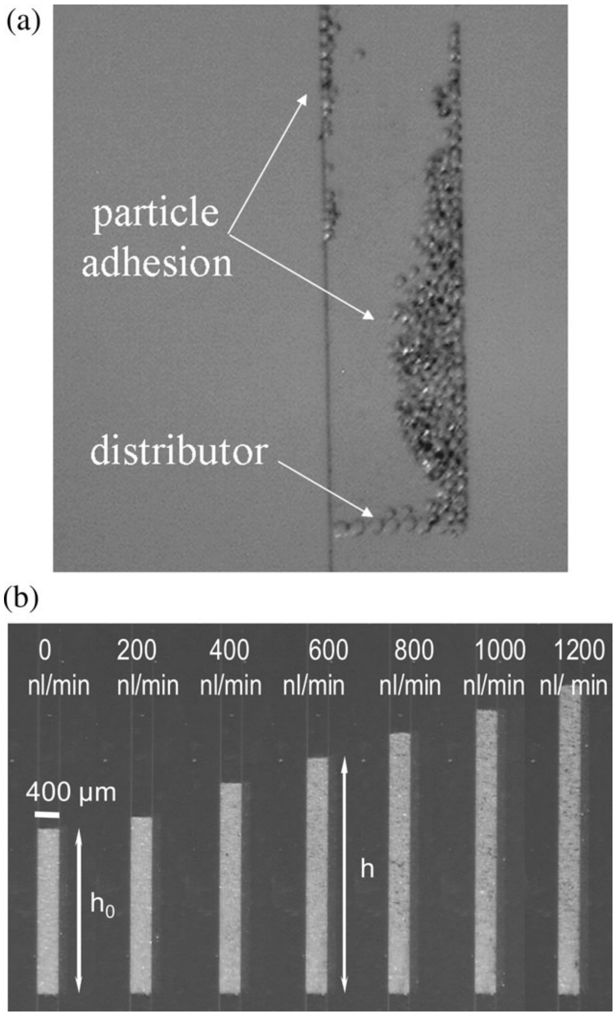

Figure 2. Optical micrographs of the microfluidized bed when (a) deionized water and (b) ethanol are used as the fluidizing medium.

Initial height $h_o = 3.215 \pm 0.015$ mm for ethanol fluidization (see Supporting Information, Video S1 and S2 online for greater detail).

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 (~8 based on hydraulic diameter of microchannel, $D_h = 243.5$ μm).[18] At flow rates at or below 200 nL/min, minor particle leakage through the distributor was observed (see Supporting Information, Video S2); 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.

ImageJ[23] 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 Figure 3 for two defluidization

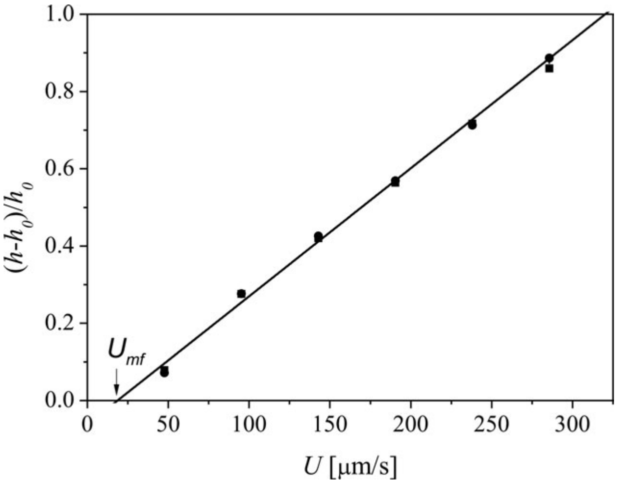

Figure 3. Relative bed height $(h - h_o)/h_o$ as a function of ethanol superficial velocity, U, for two experimental runs: squares (first run, h_o = 3.335 mm) and circles (second run, h_o = 3.215 mm).

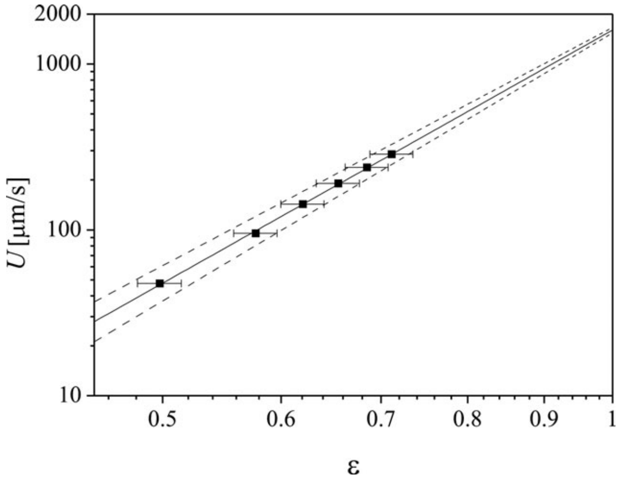

Figure 4. Microfluidized bed expansion characteristics of 30-μm glass particles inside a 400 × 175 μm² microfluidics channel.

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).

experiments. The fact that the data from the two experiments are essentially identical indicates the fluidization behavior 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 Figure 3 to the packed bed height. Although this introduces some uncertainties, the estimated minimum fluidization velocity of $U_{\mathrm{mf}} = 17.25 \pm 1.35$ μm/s falls within the range of 18.3 ± 3 μm/s predicted by the Ergun equation[24] using an estimated packed bed voidage* of $\varepsilon_{\mathrm{mf}} = 0.46 \pm 0.02$. This somewhat higher bed porosity than the value typically used for packed beds in fluidization experiments, $\varepsilon_{\mathrm{mf}} = 0.4$,[25] is a consequence of the significant effect that the wall has on the packing here relative to typical fluidized 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 Figure 3, the bed expansion characteristic for the microfluidized bed shown in Figure 4 was estimated using

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

where $\varepsilon_{\mathrm{mf}} = 0.46 \pm 0.02$ (see footnote*). The bed expansion data in this figure is well described by the well-known empirical Richardson–Zaki equation,[26] which can be expressed as

$$U = kU_t\varepsilon^n \tag{2}$$

where U_t is the particles settling velocity, k a parameter that was set to unity in the original work of Richardson and Zaki[26] 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 Figure 4 an exponent value of $n = 5.07 \pm 0.30$, which is clearly greater than that of the original Richardson and Zaki[26] work, but closer to a value of 4.9 obtained by Fan *et al.*[27] 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 ± 60 μm/s, which is almost double the theoretical Stokes law terminal velocity, U_t = 836 μm/s. This suggests that for the microfluidized bed considered here $k = 1.91 \pm 0.07$, which is in stark contrast with macroscopic experiments where $k \leq 1$ in general,[25] with only Fan *et al.*[27] 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 defluidized zone near the distributor may also play some part, at least in our experiments. Fan *et al.*[27] 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.

Conclusions

Fluidization of ~30 μm particles within a microchannel of 175 × 400 μm² 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 microchannels. Comparison of the behavior observed in this microfluidized bed system with the conventional equations for fluidization shows that these equations are a good starting point for the design of microfluidized beds. The Ergun equation[24] is applicable for evaluating the minimum fluidizing velocity provided the effect of the small particle/microchannel

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*Porosity is based on weighing of settled particles inside square glass capillaries (200 × 200 microns). Four different size microparticles were used giving corresponding channel-to-particle ratios of 5.2, 5.8, 6.6, and 7.5. We extrapolated these results to channel-to-particle ratio of 8 to obtain estimate of porosity inside our microchannel. We used double the absolute error of porosity measurement to take into account extrapolation error and difference between microchannel and capillary (aspect ratio and material).

diameter on the packed bed voidage is accounted for. The original empirical Richardson and Zaki[26] correlation, Eq. 2, also describes the microfluidized 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 microfluidized bed systems.

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