Interplay of surface forces and wall effects at a boundary of micro-fluidization

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Abstract: Micro-fluidized beds represent a novel means of significantly enhancing mixing, mass and heat transfer under the low Reynolds number flows that dominate in microfluidic devices. Major differences from their classical macro-scale counterparts are the critical importance of surface forces, which can even prevent fluidization, and almost unavoidable wall effects due to small bed size. Our experiments show interesting fluidization behavior at the boundary of micro-flow as a result of interplay between surface forces and wall effects.

Introduction

Microfluidics [1] is the science and technology of processing and manipulation of tiny volumes of fluids in channels of sub-millimeter dimensions. This research area holds promise in disparate fields ranging from automation of chemical analysis [2] and medical diagnostics [3] through to process intensification [4]. Fluidized beds have long been used at the macro-scale to enhance mixing, heat and mass transport. Our recent experimental work [5] has demonstrated that micro-fluidized beds (μFBs) are feasible, offering the potential to not only overcome diffusion-limited mixing, heat and mass transport in simple micron-sized channels, but also provide higher sensitivity and multi-modal detection in the diagnostic context by use of micro-particles [6].

In general, the main difference between micro- and macro-scale flows is the importance of surface forces relative to volumetric forces such as gravity. Based on this criterion, widely asserted boundary between two regimes is set at 1 mm [7, 8]. Not unsurprisingly, our recent work [5, 9, 10] confirmed that surface forces play an important role in μFB as well, for example surface forces can prevent fluidization in μFB in some cases. We showed that the acid-base model of van Oss, Chaudhury and Good combined with the Derjaguin approximation [11] can successfully predict the propensity of micro-particles to adhere to the walls of μFBs using common liquid fluidizing media [9, 10]. Furthermore, comparison of surface and hydrodynamic driving forces locates the boundary between micro- and macro-scale fluidization at 1 cm with stricter limit at 1 mm, the same as for microfluidics [9].

A second major issue in μFBs is the high potential for the particle-to-bed diameter ratios to be greater than 0.1, leading to significant influence of the bed walls on the packing of the particles in the bed and subsequently fluidization behaviour [5, 10]. The bed voidage in the μFB is indeed substantially higher compared to macroscale beds, leading to a significant increase in the minimum fluidization velocity [5, 10]. The bed expansion behavior also varies with the particle-to-bed ratio confirming strong wall effects as in original Richardson-Zaki correlation for viscous flow [12]. However, our preliminary experiments indicate that the Richardson-Zaki exponent, n, increases significantly in a linear manner with the particle-to-bed diameter ratio only when the ratio exceeds 0.1 [10]. We performed new experiments with glass micro-particles and water as a fluidizing medium in a 1 mm² Perspex μFB. Our experiments show that both surface forces and wall effects influence fluidization and de-fluidization behavior at this boundary of a micro-fluidization according to our previous mentioned study.

Experimental setup

The μFB was made by milling 1 mm x 1 mm channel into a Perspex block fitted with a distributor. The distributor was 1.5 mm thick, porous polyethylene sheet with mean pore size of 21μm (SPC Technologies Ltd, UK). Fig. 1 gives schematics of the experimental set-up which consisted of syringe pump (AL-4000, WPI Inc., US) to pump water as a fluidizing medium and Euromex Nexus trinocular digital microscope fitted with a USB digital camera (JB Microscopes Ltd, UK) for visualization of μFB expansion behavior. The images were stored on a PC for offline analysis. We used as-supplied soda lime glass microspheres of three different diameters, d = 35 ± 3, 58 ± 5 and 115 ± 9 μm, whose density is ρ = 2520 kg/m³ (Cospheric LLC, US).

![Fig. 1. Schematic of experimental setup for the top-view flow visualization.](image)

Results and discussion

In the acid-base approach developed by van Oss, Chaudhury and Good [11], the surface tension, γ, of the liquids and solid surfaces is expressed as a sum of an apolar, or Lifshitz-Van der Waals, component, γ_vdW, and a polar, or Lewis acid-base, component, γ_ab, that is in-turn expressed as a product of a Lewis acid, electron-donating, component, γ_d, and a Lewis

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base component $\gamma'$. Following the Dupre equation [11], the free energy of interaction, $\Delta G_{int}$, between two different solid surfaces immersed in a fluid can be evaluated using these components (see [9]). If the evaluated free energy is non-negative, the solids repel in the liquid whilst they attract if the free energy is less than zero.

The surface tension parameters for the water, wall (PMMA) and particle (glass) solid surfaces used in our new micro-fluidization experiments are sourced from various publications (4 and 3 set of values for PMMA and glass respectively, see [9]). The free energies of interaction between glass particles and PMMA walls in water were evaluated using all combinations of these values to obtain the averages of $-3.44 \text{ mJ/m}^2$ and standard deviations of $8.27 \text{ mJ/m}^2$. This suggests that glass microparticles have a small propensity to adhere to a PMMA walls surfaces in the presence of water. Our experiments confirmed this as some glass particles adhered to walls above the bed after fluidization was stopped and the bed collapsed. Although the free energy allows identification of the propensity for particle adhesion to a surface, the ratio of the adhesion and drag forces experienced by a particle in $\mu$FB is required to understand if adhesion to the bed wall will in fact occur. We estimated adhesion force using the Derjaguin approximation and the drag force evaluated from buoyant weight of particles to obtain ratios of these forces [9]. This ratio is a strong function of particle diameter as adhesion forces scale with radius while drag forces scale with cube of radius.

In our previous study the surface forces were 3 to 5 orders or magnitude larger than the estimated drag forces making fluidization impossible due to the particles adhering to the walls of the microfluidized bed [9]. However, weaker adhesion forces and bigger particles compared to our previous study [9] resulted in drag forces comparable to adhesion forces, specifically ratios of 285, 104 and 26.5 for 35, 58 and 115 $\mu$m particles respectively. Subsequently, no complete de-fluidization was observed and we were able to achieve smooth fluidization for each size of particles.

Fig. 2 shows the relative bed height as a function of fluidization velocities normalized by the theoretical minimum fluidization velocities for three different sizes of particles. In all cases the onset of fluidization is postponed which indicates that the adhesion forces between particles and the bed walls must be overcome before the fluidization starts. Interestingly once these forces are overcome, the achieved expansion of the bed is more to the point which would be expected had fluidization started at theoretical minimum fluidization velocity as can be seen by sudden jump in expansion curves especially for smaller 35 and 58 $\mu$m particles.

The experimental fluidization velocity is approximately 2, 5 and 7 times bigger then theoretical for 115, 58 and 35 $\mu$m particles respectively. Although this corresponds to the level of surface forces importance, it is not completely proportional to surface/adhesion forces ratios, but rather scale with the product of force ratio and the particle-to-bed diameter ratio as can be seen in inset of Fig. 2 (coefficient of determination 0.96). Therefore, both surface forces and wall effect influence fluidization/de-fluidization behavior of $\mu$FBs at the boundary of micro-fluidization.

**Fig. 2. Relative bed height of $\mu$FBs as a function of fluidizing velocity in terms of multiple of theoretical minimum fluidization velocity, $U_{mf}$. Inset: Ratio of experimental and theoretical $U_{mf}$ versus product of force ratio and particle-to-bed size ratio.**

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**5 References**