Etude expérimentale de la migration en microfluidique

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Résumé:

Lorsqu'une suspension semi-diluée de particules non-colloidales s'écoule dans un canal, la concentration est hétérogène car les particules migrent vers les régions peu cisaillés de l'écoulement. Nous revisitons ce phénomène dans des écoulements microfluidiques confinés, en comparant des suspensions de particules rigides avec des émulsions constituées de gouttes de très grande viscosité. Les fractions volumiques vont de 20% à 40%. Nous utilisons une approche expérimentale originale permettant la mesure simultanée des profils de concentration et de vitesses, et de la vitesse relative entre les particules et le fluide suspendant. Les deux systèmes montrent des profils de concentration similaire. Cependant, il existe une grande différence sur la stabilité aux temps longs de l'écoulement. Pour les particules rigides, nous observons une diminution continue du débit, alors que l'écoulement reste stationnaire pour les émulsions. La mesure des vitesses relatives montre que pour les émulsions, les gouttes se déplacent plus lentement que le fluide.

Abstract:

When a semi-dilute suspension of non-colloidal particles flows in a channel, the concentration of particles becomes inhomogeneous because particles migrate towards low sheared regions of the flow. In this work, we revisit this phenomenon in confined microfluidic flows by comparing suspensions of rigid particles with emulsions of high viscosity droplets, both having a volume fraction between 20% and 40%. We report an original experimental approach that allows simultaneous measurements of concentration and particle velocity profiles, and relative velocity between the fluid and particles. Both systems exhibit similar concentration profiles due to the transverse migration. However, there is a striking difference between the two, concerning the long time stability of the flow. With rigid particles, the flow rate continuously decreases within an hour, whereas it is steady for emulsions. The measurement of relative velocities reveals that at least for emulsions, the particles travel slower than the suspending fluid.

Mots clefs: **particle migration, microfluidics, semi-dilute suspensions, confocal microscopy.**

1 Introduction

The shear-induced migration of particles, which could be cells, droplets, rigid spheres etc… across streamlines, has been a subject of intense research over the past few decades. Particle migration leads to a non-uniform concentration profile that strongly affects the flow properties of the suspension. As an example, in microcirculation the so-called Fahraeus-Lindqvist effect leads to an apparent reduction of blood viscosity because of the exclusion of red-blood cells from the capillaries walls. In channel flow of suspensions of hard-spheres, it has been observed that particles do accumulate in the channel center, leading to blunted flow profiles [9]. The migration of particles is caused by the gradient in shear-rate, the stress-induced by contact forces or gravity force. In the previous two cases the particles reach zone of low shear-rate where the rate of collision between particles becomes minimum. Several models have been proposed to describe particle migration in suspensions. As for suspensions of rigid spheres, the most popular model is the suspension balance model (SBM) of Nott and Brady [10]. It attempts to describe particle migration from a rheological point of view in which the divergence of particle phase stress drives the shear-induced drift velocity of particles. Lhuillier [8] has recently pointed out some important considerations on shear- and stress-induced migration, based on a more rigorous treatment of the stress decomposition. These new theoretical insights on the migration problem underline the fundamental role of non-hydrodynamic forces, and bring some open questions especially in confined flows, where the width of the channel is typically less than a few tens of particles. In the work of Cunha and Hinch a model of rigid non-colloidal spheres in a simple shear flow is presented. They studied by simulations the effect of particle roughness on shear-induced dispersion. They present that irreversible migration of particles across streamlines in a dilute suspension depend upon their asperities on surface [4].

When particles are soft and could deform such as in the case of droplets, a drift flux due to particle deformation needs to be considered [14]. However, as compared to the case of rigid particles, rather few experimental works have been reported for emulsions.

All the major experimental works on migration have been performed in a rheometer. The pioneering work of Leighton and Acrivos brought very important observations [7]. Analyzing the migration phenomenon in a Couette cell, they show that particles migrate towards low shear zones of the flow, consistently with successive observations in channels [9].

The development of a variety of microfluidic applications dealing with complex flow at different volume fractions brings the necessity of a deep knowledge of the migration phenomena and flow stability in microchannels. There are few experimental results, to the best of our knowledge, concerning suspensions flowing in confined systems. As an example in the work of Isa et al. [5] a concentrated colloidal suspension in a micro-channel has been studied. They reported an unsteady flow that exhibits important velocity fluctuations. Such a behavior is not completely understood, but at first indicates that a steady state flow might not exist also in the simplest geometry as a square straight channel. The high volume fraction of the colloidal suspension poses some difficulties on migration visualization.

In the light on the new theoretical assumptions, our experimental work aims to investigate the role of *contact forces* by using two different suspensions: a suspension of rigid particles and an emulsion of high viscous droplets. Indeed, with the second system, contact forces are negligible since the surfaces are flat down to the nanometer scale. The high viscosity ratio of the droplet to the suspending fluid should also prevent important deformations. We aim to compare the migration and flow stability between these two systems.

Finally, as recent theories suggest that additional confinement effects need to be considered, we aim in studying confined flows, in order to decouple the intricate role of non-hydrodynamic interactions and *confinement* induced hydrodynamic forces. *Long-term stability* and *clogging issues* are also taken into account.

Since the *relative velocity* is one of the key aspects of the revisited theoretical approaches we believe also that it is a key parameter for flow stability. Up to our knowledge, the relative velocity between particles and suspending fluid has never been measured directly, though its existence in the transverse direction has been proved by migration observation. If there is a velocity difference in the transverse direction, one might also expect that there is also a velocity difference in the flow direction. This is indeed predicted by the revised SBM [11]. In this work, we propose an original experimental technique to measure this velocity difference. The fluorescence photobleaching under flow [3] is used to measure the velocity of the suspending fluid where a molecular dye is dissolved. Simultaneously, particle tracking technique [2] is employed to measure the particle velocity. We demonstrate that a *velocity difference* in the *flow direction* exists in the case of the emulsions where the velocity of the buffer fluid is higher than droplets velocity.

2 Materials and methods

2.1 The suspensions

Our model emulsion composition consists of a dispersed phase of poly(dimethylsiloxane) (viscosity 60.000 mPa.s) droplets in a continuous phase of glycerol (49,25%*w*), water (49,25%*w*) and the nonionic surfactant Tergitol NP10 (1,5%*w*, nonyl phenol ethoxy 10). The emulsion has been premixed at a volume fraction of 85% by gentle mixing to avoid the creation of small droplets. In order to obtain an emulsion with the desired size of droplets, an ad-hoc concentric cylinder Couette geometry has been used to break the droplets with a controlled steady shear [12]. The emulsion has been characterized by a Malvern Mastersizer; it has droplets of mean diameter 6.208 μm with a polydispersity of 38%. We have used a controlled-strain rheometer equipped with a cone-and-plate geometry to measure the viscosity η_e of the emulsion at different volume fraction. The emulsion viscosity follows the empiric law $\eta_e = \eta_f(1 - 1.25\Phi)^{\frac{-5}{2}}$, where η_f is the buffer viscosity.

The suspension of rigid spheres has been obtained by suspending polymethylmethacrylate (PMMA) spheres (Spheromers, CA6) in a mixture of water (10%) and thioglycerol (90%). The particles have a mean diameter of 6 µm with a polydispersity of 5%.

Both emulsions and suspensions are matched in density and refractive index at room temperature and they are florescent labelled by adding in the buffer solution a mixture of fluoresceine isothiocyanate (FITC) at 0.1 x 10^{-3} mol L^{-1} and NaOH at 2.5 x 10^{-3} mol L^{-1} to reach a pH of 7.

2.2 Confocal microscopy and experimental parameters

The capillaries (see figure 1) have a rectangular section of s_1= 40 µm x400 µm and s_2=50 µm x500 µm and are made of borosilicate glass (Vitrocom) with untreated smooth walls. The flow is driven by a pressure controller (Fluigent).

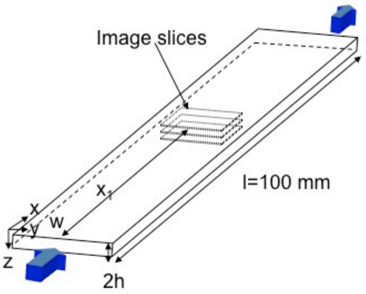

FIG. 1 – Scheme of the flow geometry. In the picture is indicating the image slices taken with the confocal microscopy.

Building up a complete picture of colloidal flows requires dynamical information at the single particle level. We measure three local quantities simultaneously, as a function of *z*: the particle volume fraction, the particle velocities and the suspended fluid velocity. Laser scanning confocal microscopy (Observer.Z1 LSM 5 Live, Zeiss) is used to acquire images at a frequency of 60 Hz in a fixed area of 167.94 µm x 167.94 µm. In the imaging window it is created a rectangular region of size of the order of 8 µm which is exposed to an high power laser intensity (100 mW) for a time of about 500 ms causing the bleaching of the dyes in that region. After, this bleaching step, the laser intensity is kept homogeneous at much lower power intensity so that we can track the displacement of the 'bleached line' as a function of time. The analysis procedure to recover the buffer velocity is similar to the one detailed in [3], except that the particles are masked before the tracking of the bleached line, and that the bleaching step is repeated every 20 frames to evaluate the mean velocity of the suspending fluid and the uncertainty of the measurement.

An important parameter for the characterization and quantification of particle migration is the volume fraction (Φ). The precise measurement is usually a difficult task [13]. In this work we experimentally measure it. The volume fraction is defined as the fraction of the total volume *V* that is filled by *N* particles of radius a, $\Phi = \frac{4}{3}\pi a^3 \frac{N}{V}$. Particle identification is performed by means of the circular Hough transform. Moreover this method allows to determine the particle radius a from the images with sub-pixel accuracy and to reject out of focus particles. We determine the volume fraction associating the number of particles in a volume, which is defined by the optical system. This volume is calibrated thanks to particle counting in droplets of known volume fractions. The employed method leads to a precision of about 1%.

Finally, using the particle positions, single particle tracking technique is used to calculate the trajectories in two dimensions. We use a modified version of the particle tracking algorithm of Crocker and Grier [2]. From the particle trajectories, it is straightforward to obtain the mean velocity and the velocity fluctuations as a function of z.

3 Results

3.1 Stability of the steady-state

In microfluidic applications involving the flow of complex fluids, it is of primary importance to reach a stable and controlled flow. For this reason the flow has been monitored during about 1 hour. Figure 2 displays the time evolution of the mean velocity of both suspensions and emulsions at different volume fraction in a channel of width 50 μm. There is a striking difference between the two systems. Emulsions do exhibit a well-defined steady state. On the contrary, the flow rate of suspensions of rigid particles slows down in time. This very slow time evolution is puzzling, and does not seem to evolve towards a reproducible steady state: during some experiments, the system evolves towards clogging. We have checked experimentally that channel clogging is not due to inlet or outlet clogging by employing in the set up large inlet and outlet so that they can be considered open. A possible interpretation may reside in particle migration in the direction of the flow as predicted by the revised SBM [11]. It is proved that the evolution of this slowing down can conduct to the formation of clogs. To go deep in this study it is necessary to analyze the collective dynamics of hard-spheres and droplets in a confined flow and how the deformability and surface characteristics can affect the flow properties.

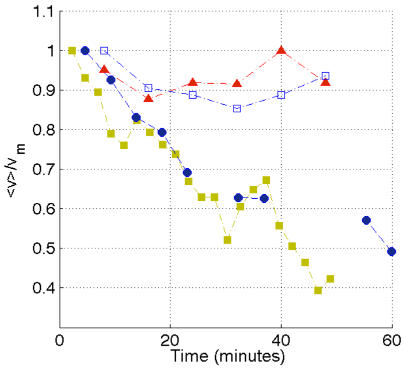

FIG. 2 – Time evolution of the mean velocity in a 50 μm thick channel at z=25 μm, for various suspensions and emulsions at different pressure drops: hard-sphere suspensions driven at 30 mbar for Φ=40% (full squares), and Φ=30% (circles), emulsions at Φ=40%, for a pressure of 5 mbar (triangles) and related buffer fluid velocity (light squares). The mean velocity is normalized by the maximum velocity.

As a recent study points out, there is an intrinsic difference between a linear array of rigid and deformable particles in a Poiseuille flow [6]. We believe that the complexity of the system increases if we consider multiple arrays of particles disposed in a 2D space.

3.2 Spatial variation of the flow properties

In order to get some additional insights on the slowing down of the flow occurring with suspensions of rigid particles, we check the spatial evolution of the concentration and velocity profiles after 1 hour of experiments. Figure 3 shows these profiles in two different locations of the channel along the x-axis. The two profiles do not superimpose, the velocity is flatten in the center of the channel as x increases, similarly to the concentration profiles. The latter also exhibit a small increase in the mean concentration. Thus the particles accumulate and spread towards the wall when travelling in the channel. It can however explain why the flow exhibits systematically a slowing down as a function of time. The time and space scale of this secondary migration are surprisingly very long. It is interesting to notice that, in the case of suspensions of rigid particles, the velocity of particles fluctuates in time, at a time scale of a few seconds. The velocity fluctuations are of the same order of the mean velocity. This observation might also be a consequence of the particle accumulation, but may also be linked to the presence of secondary currents [15] attributed to the second normal stress difference in flowing suspensions [1].

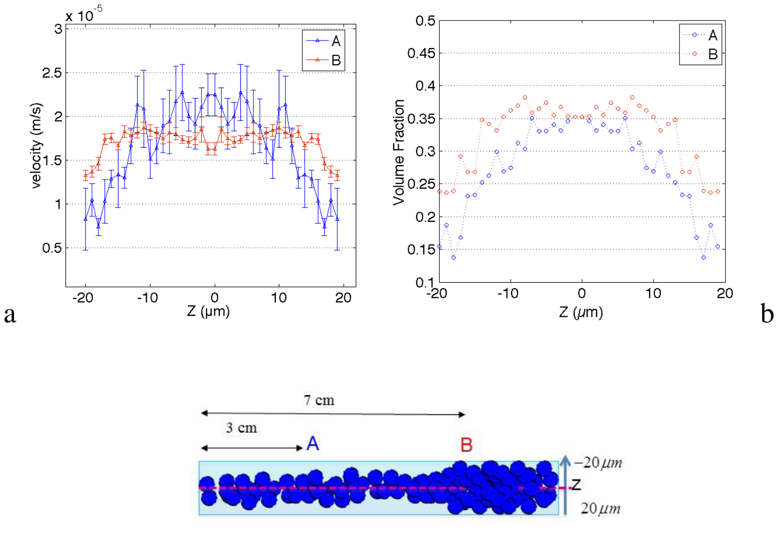

FIG. 3 – (a) Velocity and (b) concentration profiles along x in a 40 μm thick channel for a suspension at Φ=40% and ΔP=10 mbar for Δt>1 hour. The two profiles are taken at 3 cm (A) and 7 cm (B) from the inlet. At the bottom is schematized a possible scenario: the rigid particles accumulate and spread when travelling in the channel.

3.3 Velocity differences

As explained in the introduction, the mechanisms leading to migration should also induce velocity difference in the flow direction. Such a velocity difference indirectly is suggested by the observation of particle accumulation during the flow of hard-spheres. In order to describe this migration in the flow direction, we measure simultaneously the velocity of the suspending fluid and that of the particles, as detailed in the section devoted to the experimental methods. We first apply this method to the flow of emulsions because it exhibits a well-defined steady state.

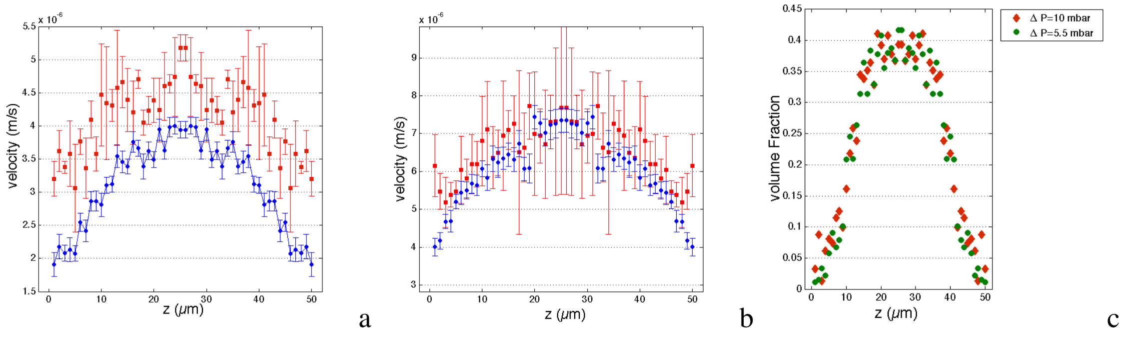

FIG. 4 – Flow profiles of buffer (red squares) and droplets (blue circles) in a 50 μm thick channel at Φ=30% for (a) ΔP=5.5 mbar and (b) ΔP=10 mbar, (c) volume fraction profiles.

The respective velocity profiles of the particles and of the suspending fluid are plotted in figure 4. It is clear from these results that the buffer fluid travels faster than the emulsion droplets for lower pressure. This difference is on the order of about 1.5 μm/s, and is significantly greater than the measurement uncertainty.

As we can see the velocity difference between the droplets and the buffer fluid decreases when increasing the pressure drop. Similar experiments are currently conducted with the suspensions of rigid spheres and preliminary results lead to a similar trend. Existing theories need to be further developed in order to interpret these results.

It is worth to notice that the volume fraction profiles do not vary with the applied shear (figure 4(c)), in agreement with the SBM description of the migration.

4 Conclusions

In this work, we have studied experimentally the flow of semi-dilute suspensions and emulsions in a microfluidic rectangular channel. We report direct simultaneous measurements of volume fraction and velocity profiles, and a new method for measuring velocity differences between particles and suspending fluids. The preliminary results are consistent with previous results concerning the migration, but also reveal some unexpected important differences between emulsions and hard-sphere suspensions. A very slow particle accumulation and successive spreading is observed in the confined flow of rigid particles while the flow of emulsion droplets remains stable temporally and spatially with no important fluctuations. This suggests that the contact forces and interface properties of particles are fundamental especially in strongly confined system also in the case of semi-dilute regime. Moreover the measure of fluid and droplets velocity difference gives evidence that there exists a significant relative velocity in the flow direction, a feature that has not been deeply investigated yet. Perspectives of this work will deal with more systematic investigations of velocity differences and additional comparisons between emulsions and hard-sphere suspensions at different polydispersity.

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