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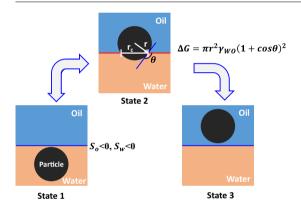
Dynamics of rigid microparticles at the interface of co-flowing immiscible liquids in a microchannel



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ABSTRACT

We report the dynamical migration behavior of rigid polystyrene microparticles at an interface of coflowing streams of primary CP₁ (aqueous) and secondary CP₂ (oils) immiscible phases at low Reynolds numbers (Re) in a microchannel. The microparticles initially suspended in the CP1 either continue to flow in the bulk CP₁ or migrate across the interface into CP₂, when the stream width of the CP₁ approaches the diameter of the microparticles. Experiments were performed with different secondary phases and it is found that the migration criterion depends on the sign of the spreading parameter S and the presence of surfactant at the interface. To substantiate the migration criterion, experiments were also carried out by suspending the microparticles in CP2 (oil phase). Our study reveals that in case of aqueoussilicone oil combination, the microparticles get attached to the interface since S < 0 and the three phase contact angle, $\theta > 90^{\circ}$. For complete detachment of microparticles from the interface into the secondary phase, additional energy ΔG is needed. We discuss the role of interfacial perturbation, which causes detachment of microparticles from the interface. In case of mineral and olive oils, the surfactants present at the interface prevents attachment of the microparticles to the interface due to the repulsive disjoining pressure. Finally, using a aqueous-silicone oil system, we demonstrate size based sorting of microparticles of size 25 μm and 15 μm respectively from that of 15 μm and 10 μm and study the variation of separation efficiency η with the ratio of the width of the aqueous stream to the diameter of the microparticles ρ .

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1. Introduction

Separation, focusing and enrichment of particles has profound applications in chemical and biological processing including sample preparation, disease diagnosis and water treatment [1-3]. Use of microfluidic techniques for the manipulation of particles have distinctive advantages in terms of lower sample volume, continuous separation and processing of samples [4]. Continuous separation of particles based on lateral migration is of much interest since the discovery of Segre-Silberberg effect [5,6]. Particles moving in an inertial flow (Reynolds number Re > 1) displace radially and occupy definite positions based on the morphology and physical properties of the particles. The equilibrium positions attained by the particles is governed by the wall induced lift, shear gradient lift, elastic lift and deformability induced lift forces [5,7–11]. In low Re flows (Re < 1), rigid particles do not experience any lift force and tend to move along their stream lines [10,12]. However, deformable objects such as cells, vesicles and droplets experience non-inertial lift force due to their deformable nature, morphology and orientation of the objects with the flow direction. Deformable objects such as droplets [10,12,13] and vesicles [14-16] initially located at an offset to the channel centreline, in shear flow condition at low Re exhibit lateral migration and move toward the channel centreline (zero shear). Using the underlying hydrodynamics of the non-inertial lift force, focusing, sorting and enrichment of droplets and cells were successfully demonstrated [17].

Study of migration of droplets from one continuous phase to another continuous phase in a co-flowing stream of miscible liquids have been studied [18]. The droplets initially present in a higher viscosity phase migrate into the lower viscosity phase, due to the presence of zero shear stress region in the lower viscosity phase. Selective interfacial migration of droplets from a primary liquid into a secondary liquid in case of co-flowing streams of two immiscible liquids was also investigated. In a co-flowing two-phase system, an additional force, interfacial tension force, becomes significant at lower length scales which compete with the non-inertial lift force to enable selective interfacial migration of droplets based on size and viscosity [19].

Migration of rigid particles in a single phase or two-phase flow at low Re is not much investigated since such particles do not experience any lift force. However, presence of rigid particles at static fluid-fluid interfaces (i.e. fluids are not under shear flow) have been studied in recent years since such studies have potential applications in the stabilization of emulsions [20-22]. Although the size of the most colloidal particles (proteins and fat) used for the stabilization of emulsions is in the range of 1.0 nm to 1.0 um. 10 um latex beads can also be used for the emulsion stabilization [22,23]. Colloidal particles have a natural tendency to get attached to liquid-liquid and air-liquid interfaces thereby reducing the interfacial energy of the system [20,24]. Spontaneous transport of rigid sulfate-treated polystyrene particles of diameter 1.0 µm and 4.0 μm from water phase to ionic liquid phase is reported [25]. The liquid-liquid interface in the study was static and the transport of microparticles across the interface, despite the strong adhesive force at the interface, is because the ions present in the ionic liquid phase which cover the particles completely and favor migration. This strong adhesion of particles at interface has been utilized to stabilize water-in-water as well as oil-in-water emulsions [20].

It is thus evident that most of the previous studies on rigid microparticles are carried out in the inertial regime and recent studies are performed with static interfaces. Here, for the first time, we study the behavior of rigid polystyrene microparticles at the interface of co-flowing immiscible liquid streams at low *Re*. The dependence of the interfacial migration of microparticles on the interfacial properties of the microparticles and the liquids is

studied using different oils and aqueous glycerol as the immiscible phases. The role of perturbation of the interface on the detachment of microparticles from the interface is discussed. Size based sorting of microparticles is demonstrated and the effect of the ratio of the width of the sample stream to the size of the microparticles on the sorting efficiency is determined. First, a brief description of the device and the theoretical background are presented. Next, materials and methods and the experimental setup is outlined. Finally, the experimental results are presented and discussed.

2. Device description and theoretical background

Our experimental studies were performed using two different microchannel designs: a simple Y-channel and a flow-focusing channel, as shown in Fig. 1. Using Y-channel, two different sets of experiments were carried out, first rigid polystyrene microparticles are suspended in the aqueous phase and their behavior at different immiscible oil interfaces is studied. Next, the microparticles are suspended in different oil phases and their behavior at the aqueous interface is studied. In the first set of experiments, the microparticles are suspended in aqueous liquid and infused into the channel as the primary continuous phase CP₁. An immiscible oil is infused as a parallel co-flowing stream alongside the aqueous phase as the secondary continuous phase CP2. The minimum stream width a of the aqueous phase is non-dimensionalized with the width w of the channel in case of Y-channel and with the width of the throat in case of flow-focusing channel, which is denoted as $\alpha = a/w$. The non-dimensional stream width α is a function of the flow rate ratio r of the continuous phases, CP_1 and CP_2 , $r = Q_{CP_1}/Q_{CP_2}$, viscosity ratio of the continuous phases and the channel cross-section [19,25]. The rigid micrometer sized particles do not experience any lift force and thus continue to travel along the streamlines passing through their center of mass [10,12]. When the width a of the aqueous stream CP_1 is of the order of the size (diameter d) of the microparticles, interaction between the microparticles and the aqueous-oil interface occurs.

When a particle suspended in a primary phase (CP₁) interacts with a secondary phase (CP₂), the wetting behavior is governed by the spreading parameter *S*. In the present system, when a microparticle initially suspended in aqueous phase interacts with an oil phase, the oil-side spreading parameter is defined as [26,27],

$$S_o = \gamma_{sw} - (\gamma_{so} + \gamma_{wo}) \tag{1}$$

where γ_{sw} is the surface energy of the solid microparticle with the aqueous phase, γ_{so} is the surface energy of the solid microparticle with the oil phase and γ_{wo} is the interfacial tension between the aqueous and the oil phases. The particle makes a three phase contact angle of θ measured through the aqueous phase as shown in Fig. 2. In this case, the force balance at the contact line can be written as, $\gamma_{so} = \gamma_{sw} + \gamma_{wo} cos\theta$ and when combined with Eq. (1) gives,

$$S_o = -\gamma_{wo}(\cos\theta + 1) \tag{2}$$

Similarly, water-side spreading parameter is defined as,

$$S_{w} = \gamma_{so} - (\gamma_{sw} + \gamma_{wo}) \tag{3}$$

Eq. (3) when combined with the force balance at the contact line gives,

$$S_{w} = \gamma_{wo}(\cos\theta - 1) \tag{4}$$

For attachment of particles to the interface, both the oil-side and water-side spreading parameters should be negative ($S_o < 0$, $S_w < 0$). If the oil-side spreading parameter S_o is zero or positive ($S_o > 0$), the microparticle should migrate completely into

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