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Water-in-oil emulsification in a microfluidic impinging flow at high capillary numbers

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ABSTRACT

The formation of water-in-oil disperse flows is investigated in a microfluidic device. The liquid–liquid flow mainly differs from these presented in the existing literature in its high capillary number (between 3 and 14). The pressure-driven high flow rate, the viscous oily continuous phase, and the head-on impinging flow contribute to oppose a maximal shear force to the interfacial tension. Interfacial tension effects are significant at scales smaller than the capillary length within microfluidic devices, but not dominant in the present work. Moreover, the impinging channel is designed to deliver a low water fraction. As a result, the obtained water-in-oil emulsion is finely dispersed. A channel geometry is selected among three different items, with a physical explanation of its observed finer emulsification. In comparison with experimental data, two complementary approaches are used to model the Ohnesorge number from fluids and flow parameters. Finally, an in situ optical diagnostic using polarized laser light provides a precise knowledge of the emulsifying area and the overall flow pattern within the microchannel.

Introduction

Liquid–liquid microfluidic flows have drawn considerable attention in the last decade Zhao and Middelberg (2011). These flows can release an emulsion, i.e. a metastable dispersion of one liquid in another immiscible Engl et al. (2008). By slowly generating one droplet at a time, microfluidic devices are able to produce highly monodisperse streams of droplets, with a precise control over their size Engl et al. (2008). Numerous applications benefit from improvement of heat and mass transfer with increased surface to volume ratios, and from droplets kept apart from each other without coalescence Burns and Ramshaw (2001) and Song et al. (2003). These applications include biomedicine Jakeway et al. (2000) or protein crystallization Li et al. (2006). Other applications are in the field of energy engineering and oil industry. In this extent, micro-explosion of water-in-oil emulsified fuel drops Tarlet et al. (2009, 2014) and Mura et al. (2010) is known to efficiently decrease pollutant emissions. These applications generally require higher flow rates and less monodispersion of water droplets. Bremond and Bibette (2012) remind that the polydispersity is governed by a competition between drop breakup and drop coalescence, that is likely to be observed at high flow rates.

The capillary number *Ca* quantifies the ratio of viscous to interfacial forces, and is known to be the most relevant number to characterize microfluidic liquid–liquid flows Zhao and Middelberg (2011) and Shui et al. (2007). In the existing litterature, microfluidic liquid–liquid flows have been extensively investigated in a low range of capillary number, with *Ca* between 0.001 and 0.1 Xu et al. (2006), Xu et al. (2006), Zhao and Middelberg (2011), Shui et al. (2007), Tice et al. (2004), and Zhou et al. (2006). The effects of interfacial tension σ_{w-o} become dominant over gravity g when considering a channel smaller than the capillary length, de Gennes et al. (2005).

Emulsification within microfluidic devices is mainly based on the interplay between interfacial tension σ_{w-o} and viscous shear and elongational forces Bremond and Bibette (2012), Xu et al. (2006), and Thorsen et al. (2001). Besides original microfluidic devices Sugiura et al. (2001), three main categories are known Zhao and Middelberg (2011) and Engl et al. (2008): T-junction Xu et al. (2006), Thorsen et al. (2001), and Link et al. (2004), flow-focusing and co-flowing Zhou et al. (2006). High-pressure, impinging-jet microfluidics are also experimented Mason et al. (2006). In a T-junction channel, cross-flowing Thorsen et al. (2001) and perpendicular flowing Xu et al. (2006) are orthogonal configurations, with similarities and differences from the microchannels used here.

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In the T-junction microchannel experimentally observed by Tice et al. (2004), viscous forces become dominant over interfacial tension between $Ca = 0.001$ and 0.1. In outstanding numerical works about the same kind of geometry de Menech et al. (2008) and Bashir et al. (2014), this interplay defines three distinct flow regimes named as squeezing, dripping and jetting. These three flow regimes are known to mainly depend on the capillary number, Ca, Zhao and Middelberg (2011). When the flow rate is low with Ca around 0.001 in the squeezing regime, interfacial tension dominates Tice et al. (2004) and Sugiura et al. (2001). Dripping and jetting regimes are also identified in flow-focusing microfluidics Zhou et al. (2006). When the viscous forces dominate, an interesting fact is noticed in the jetting regime at a higher value of Ca, between 0.01 and 0.1, Tice et al. (2004): A longer filament of the dispersed phase is formed before it undergoes shear induced breakup.

The field of the present experimental data shown in Fig. 1 is exposed in details in Section 'Emulsification in the 300 × 600 µm channel'. It is compared to the ranges of capillary number Ca and mean diameter of liquid–liquid dispersion D_d in T-junction microfluidic devices, from different authors. The Fig. 1 shows that the present results are roughly aligned with the relationship from Xu et al. (2006). Other authors that worked in a lower range of capillary number obtained a larger mean diameter, Zheng et al. (2004), Xu et al. (2006), Nisisako et al. (2002), Kim et al. (2014), and Salim et al. (2008). However, these microfluidic devices from other authors working at lower values of Ca generally produced droplets with a very good monodispersion.

In the present work, emulsification of a small fraction of water in oil is investigated in a higher range of capillary number (between 3 and 14) than it is usually practiced in the existing litterature. Four choices about physical properties of the fluids and flow conditions are made to emphasize that:

- A head-on collision of the continuous phase oil stream and the water stream, in which their kinetic energies are added. Indeed, liquid filaments are expected at values of Ca higher than 0.1 Tice et al. (2004). Head-on collision has been studied in other flow configurations Tanguy and Berlemont (2005) and Inamura and Shirota (2014).
- The viscosity of the continuous phase ($\mu_o = 52.2$ mPa s) is much higher than that of the disperse phase. It contributes to increase the viscous shear and elongation forces applied on disperse phase, Grace (1982).
- A smaller water inlet compared to the oil inlet, that delivers a low water fraction (4–11%). This is expected to increase the tendency to a disperse flow Brennen (2005), also referred to as a droplet flow Kim et al. (2014).
- Adding small amounts of SPAN surfactant used in some experiments allows to decrease the interfacial tension σ_{w-o}.

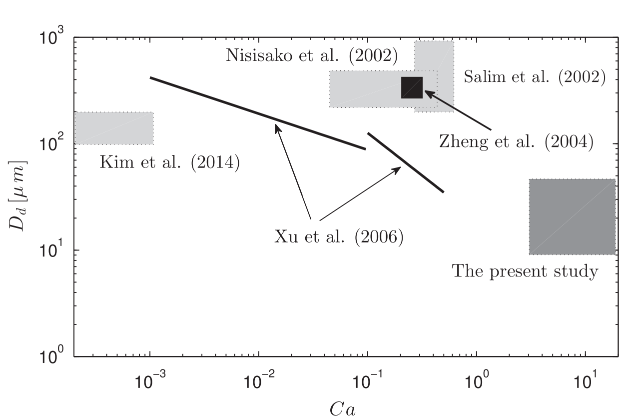

Fig. 1. Comparison of the relationship $D_d \propto (1/Ca)^{0.3}$ exposed by Xu et al. (2006) to the field of the present results, shown in dark gray. The intervals of Ca and D_d for different authors are shown, only concerning droplet flows with diameters smaller than the channel width.

To sum up, this work aims at designing devices which enhance viscous shear and elongational forces at high capillary numbers, in order to counter-balance the cohesion of filaments of water due to interfacial tension. The used microfluidic devices produce a finely dispersed water-in-oil emulsion at a flow rate (mL/min) three orders of magnitude higher than what is practiced in most of the existing litterature.

In this paper, we first experiment three different devices so as to select the best channel geometry, i.e. water to oil inlet size ratio. In this extent, the selected emulsifying microchannel is producing emulsion with the strongest cohesive force. Secondly, the selected device is investigated so as to characterize the physics of its impinging jets emulsification. The effects of fluid properties (interfacial tension, viscosity) and flow properties (flow rate) on the obtained emulsion are investigated. A dimensional analysis of emulsification is performed to predict the obtained Ohnesorge number. Moreover, a simple pressure balance downstream the impinging zone is compared to the experimental data. The purpose of these two different approaches is to give a physical explanation to the impinging flow emulsification, and to be able to predict the obtained size of the water droplets in the present case. Lastly, a polarized laser beam is mapping the channel in order to detect the presence of water droplets, especially the stretched and elongated. Such droplets or filaments are expected where intense velocity gradients are located. The knowledge obtained about the emulsifying channel consists in the size of the emulsifying area within the impinging flow, and also in the overall liquid–liquid flow pattern.

Experimental set-up and instrumentation

The microfluidic facility to investigate water-in-oil (w/o) emulsification within impinging streams is shown in Fig. 2. Two piston pumps (ARMEN-APF-100-25-1) are used for supplying water and filtered sunflower oil at a high pressure (up to 5 bars). The overall energy consumption of the emulsification process is measured through the electrical power consumed by the pumps. It ranges between 45 and 55 W, while the energy released by combustion of the present flow rate of sunflower oil ranges between 30 and 50 kW. Flow rate measurements are performed using weighing scales (Sartorius-MSE 2203, 1 Hz sampling) connected to a computer. The weighing scales do have a measurement accuracy of 10^{-3} g, which results in an uncertainty of 5% for the flow rate. The pressure is measured at the water and oil inlets using Compact Pressure Transmitters (Gems 3100 series, Pressure range 0–25 bars with an accuracy ±0.25% of the full scale, 1 Hz sampling). All connections between pumps and mini-channel are made using Fluoropolymer (FEP) tubing with internal diameter of 1.55 mm. The outlets are at the atmospheric pressure.

The flow pattern is observed with a fast CCD camera LAVision HighSpeed Star 6 equipped with the macro objective necessary to watch at the millimeter scale. The focus was made in the channel depth. The light source consists of a 50 W halogen spot located in the line of sight, beyond the transparent microfluidic device.

Design of the microfluidic devices

The geometry and design of the devices is presented in Fig. 3. It consists of crossing microchannels featuring two inlet sections and two outlet sections. The channels have a square cross section, as