

Microdroplet Formation in Microfluidic Channels by Multiphase Flow Simulation

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Abstract— Microfluidics is a growing area of micro and nanofabrication that found broad applications in molecular biology, chemistry, pharmaceuticals, synthesis, optics, and informatics, among others. Microfluidics was developed from the interaction between molecular analysis, biodefense, molecular biology, and microelectronics. Some cell fabrication processes and microfluidics experiments could be expensive, then the computational simulation of experiments is useful to establish the appropriate conditions before the fabrication. In this work, we present the microdroplet production using two immiscible liquids (oil and water), their transport, as well as the general procedure of computational fluid dynamic (CFD) simulation. We used time-dependent two-phase microfluidic flow in 2D with a “T” cell geometry with 1 μm of width. In all cases, droplet production can be achieved until a water inlet velocity limit. We observed an inverse relationship between the droplet distance and the inlet velocity. If the distance between droplets decreases, the inlet velocity increases. However, when water inlet velocity increases to reach the Weber number larger enough to overcome interfacial tension, a continuous flow of water is achieved. This occurs when $v = 0.02$ m/s of inlet water flux. We do not observe squeezing regime because $x = \text{width}_{in}/\text{width}_{out} \ll 1$, and then droplets do not obstruct the channel as it grows, neither restricts the flow of continuous phase.

Keywords—*microfluidics, multiphase flow simulation, COMSOL.*

I. INTRODUCTION

Microfluidics refers to the manipulation of fluids in channels with the dimensions of tens of micrometers. A search in the PubMed database with the word “microfluidic” shows that the number of articles related to the field has been triplicated from 1133 to 2835 articles per year, during the past ten years. It finds many applications in areas such as molecular biology, chemistry, pharmaceuticals, synthesis, optics, and informatics, among others. Microfluidics is the science and technology of systems that process or manipulate small amounts of fluids (10^{-9} to 10^{-18} liters), which reduce the use of reagents, allowing to carry out many experiments or reactions with high resolution and sensitivity, low cost and rapid analysis. Mainly, in vital areas where reactants and biological materials like nucleic acids, molecules, proteins,

enzymes, antibodies or drugs are limited to carry out parallel experiments, microfluidics is very helpful.

The microfluidics exploits both, the small physical scale where liquids are confined, and the laminar fluid phenomena developed inside the channels. There are several types of cells used in microfluidics such as open microfluidics [1], paper microfluidics [2], continuous flow chemistry [3, 4], microfluidics by capillaries [5, 6] and more recently, highly integrated microfluidic devices [7], lab on a chip [8], Micro Total Analysis Systems (μTAS) [9, 10] and organ on a chip [11-13]. In this article, we review the simulation, fabrication of microfluidic devices and its use for the microdroplet formation.

Microfluidics was developed from the molecular analysis (gas-phase chromatography, high-pressure liquid chromatography, and capillary electrophoresis), biodefense, molecular biology, and microelectronics [14].

Mainly, the incorporation of microelectronics was motivated by photolithography and the well-established silicon technology, to produce microfluidic systems at low cost and high production rate. In the beginning, some microfluidic systems were made in silicon, but its cost and opaqueness to visible light avoided the use of optical techniques for analysis. Recently, polydimethylsiloxane (PDMS) and plastics replaced the use of silicon and glass. PDMS is a hydrophobic, transparent, soft, and low reactive elastomer. When it solidifies, allows its use as imbedded systems like valves, pumps, electronic components, among other elements, that will be difficult to implement on silicon devices.

One application of microfluidics is the possibility to form microdroplets of oil in water, water in oil or a combination of layers such as w/o/w or o/w/o [6]. These microdroplets can be generated continuously, and with excellent reproducibility, not only the synthesis but also encapsulation is possible. It gives them desirable characteristics for industry, chemistry, biology and pharmaceutical applications. In this way, each drop can be viewed as a micro-reactor with

volumes from *pico* to *femtoliters*, with a volume of 4.18×10^{-15} liters per particle of $20 \mu\text{m}$ of a radius. They are usually made within a carrier fluid of oil (the carrier phase) and an inner phase of hydrophilic liquid (water, for example).

This condition of two immiscible fluids is necessary for droplet formation, and desirable being that the carrier fluid forms a barrier which avoids evaporation and confines the inner fluid inside the channel. Also, it compartmentalizes one droplet from another and avoids the residues or content from one droplet to be in contact with another. This condition can be viewed like independent experiments or reactions continually occurring in a continuous flow inside the channel.

Other physical characteristics like density, viscosity, interfacial tension and velocity of the phases play an important role, even also the geometry of the channels is essential to improve or diminish the physical desired phenomena.

Due to the dimensions of the channels and laminar flow, mixing is mainly produced by diffusion. So, specific geometries are required to achieve mixing, splitting, sorting or droplet generation [14, 15]. Different flow regimes can appear on the same channel by varying one condition such as velocity, viscosity or density of the fluids [5]. Some references describing the micro drop formation and microfluidic governing laws can be found in [15-23]. One of the standard conditions in the channels is the dripping to jetting transition inside micropipettes, first studied by Utada *et al.* [5]. They analyzed the inner and outer flow rate, viscosity ratio, surface tension, capillary number of the outer fluid, and Weber number of the inner fluid. Here, we limit our scope to the production of droplets and their transport as well as the general process of computational fluid dynamic (CFD) simulation.

II. SIMULATION

A. Computational Fluid Dinamic Simulation

The use and development of microfluidic devices are increasing, and it is expected that such devices will replace actual methods to perform chemical and biological assays [24]. However, some microfluidic devices require expensive equipment and clean room facilities for their fabrication; then computer simulation can help to reduce both, time and cost. Besides, it allows determining how geometry, selected materials and design affects the device performance, reducing the prototyping fabrication cost [25, 26]. This pathway has been used in other areas like electronics, construction, and bioengineering, where the model is visualized, numerically tested, adapted if it is necessary, and finally fabricated.

B. Microdroplet formation

The primary physical condition for droplet formation of two immiscible liquids is the tension at the interfacial region. The corresponding pressure is denoted as γ (Nm^{-1}). When this force suffers a change, it produces shape deformations at the interface, inducing flow from the lowest to the highest interfacial tension regions. Then, the interfacial tension tries to minimize the surface area to reduce the energy at the interface until a sphere shape is formed, leading to the formation of droplets of the dispersed phase into the carrier phase. When the interphase deforms and curves, it introduces

a pressure known as *Laplace Pressure*, which is different at the inside and outside of the droplet as $\Delta P = \gamma(1/R_1 + 1/R_2)$, where R_1 and R_2 are the radii of curvature of the interphase [21].

Table 1 shows some dimensionless parameters that relate physical characteristics of the fluids of interest in microfluidics. Reynolds number (Re) relates density, velocity and unit length against viscosity of the fluid. Thereby, high numbers denote turbulent flow, generally above 4000, whereas a Re number between 2100 and 4000 indicates a transition regime where the liquid start to lose its stability and rhythmic undulations over time appears. Nevertheless, below 2100 the viscous force plays a more critical role and dominates the behavior of the liquid, causing laminar flow.

In this regime, the Weber number We is small allowing the use of both, low volumes of liquids and dimensions of channels. In this case, the Bond number Bo is negligible due to its minimal contribution. By its side, the Capillary number Ca compares viscosity and interfacial tension, which are essential at a small scale. When more than one fluid is present in the microfluidic cell, it is considered the Ca contribution of the fluid with higher viscosity.

Low values of Ca indicate that interfacial tension is bigger than viscous forces. This condition generates spheres in fluids because the drop minimizes its surface energy at the interface. When viscous forces dominate, we observe more significant drops and asymmetric shapes [27].

C. Physical considerations in a multi-phase microfluidic flow

In the mechanic of fluids, the *Navier-Stokes* equation is usually used (**Eq. 1**) in the case of incompressible Newtonian fluids, described as a continuous system (**Eq. 2**). The numerical solutions are known as Computational Fluid Dynamics (CFD).

TABLE I DIMENSIONLESS NUMBERS OF INTEREST[27]

Formula	Relationship
Reynolds number $Re = \rho v \lambda / \mu$	Relates the density ρ , velocity v and unit length λ with a viscosity μ of the fluid. We study systems at $Re < 2100$, where the flux is laminar.
Weber Number $We = \rho v^2 \lambda / \gamma$	Relates the density ρ , velocity v , and length λ (inertia) with interfacial tension γ . Relates the gravitational forces produced by a difference in liquid densities ($\Delta\rho$) and interfacial tension γ . We neglected because we consider very small volumes of fluids, $Bo \ll 1$.
Bond Number $Bo = \Delta\rho g \lambda^2 / \gamma$	
Capillary Number $Ca = \mu v / \gamma$	This number compares mainly viscous forces μ and γ .

$$\rho \frac{\partial \mathbf{u}}{\partial x} + \rho(\mathbf{u} \cdot \nabla) \mathbf{u} = \nabla \cdot [-p\mathbf{l} + \mu(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)] + \rho \mathbf{g} + \mathbf{F}_{st} \quad (1)$$

$$\nabla \cdot \mathbf{u} = 0 \quad (2)$$

$$\frac{\partial \varphi}{\partial x} + \mathbf{u} \cdot \nabla \varphi = \gamma \nabla \cdot \left(\epsilon_{1s} \nabla \varphi - \varphi(1 - \varphi) \frac{\nabla \varphi}{|\nabla \varphi|} \right) \quad (3)$$

The boundary conditions give the changes at the interphase (Eq. 3) where φ , the advection variable at the water-oil interface, is 1 inside the droplet, 0 outside, and 0.5 at the interphase (Fig. 1); ε_{js} controls the thickness at the interphase, and γ is a reinitialization parameter.

We use the *Conservative Level Set method* to simulate two phases, which present some advantages over other methods such as front tracking, the volume of fluid and phase field [28-30].

III. METHODS

A. Microfluidic simulation general procedure

In this work, we used COMSOL Multiphysics® (5.4 version) to model the droplet formation of two immiscible fluids (vegetable oil and water).

The general procedure is well known and can be found elsewhere [31]. In this study, we used a time-dependent two-phase microfluidic flow in 2D. The geometry used was a “T” intersection as is shown in Fig. 2. Blue arrow indicates the inlet and flows direction of water, red arrows correspond to inlets and flow direction of oil, and black arrow the flow outlet.

We chose variables provided in the database of the software for density and viscosity of vegetable oil and water, whereas the surface tension corresponded to corn oil/water.

The geometry, materials, conditions, initial values, interphases, and wall boundary conditions were set up (typically non-slip condition for walls), giving the three main domains shown in Fig. 3. Domain 1, in blue, (Fig. 3, top) corresponds to water meanwhile domains 2 and 3 represents oil. Furthermore, (six and eight at Fig. 3, bottom) represents interphases where liquids are in contact. We ran calculations keeping fixed initial values of oil inlets (red arrows in Fig. 2) with a normal velocity of 0.01m/s, and different water inlet velocity (blue arrow in Fig. 2) from 0.001 to 0.01 m/s.

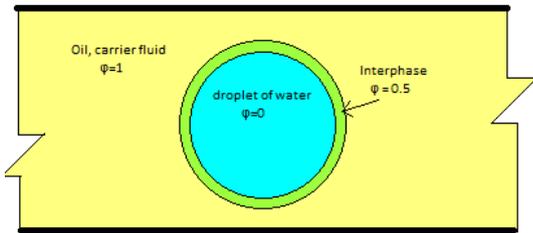


Fig. 1. Regions of water droplet formation in an oil fluid.

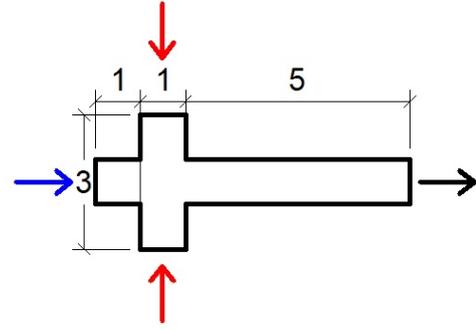


Fig. 2. Geometry and direction of incoming fluids, blue arrow indicates inlet and flow direction of water, red arrows inlets and flow direction of oil, and black arrow the outlet. Dimensions are in microns.

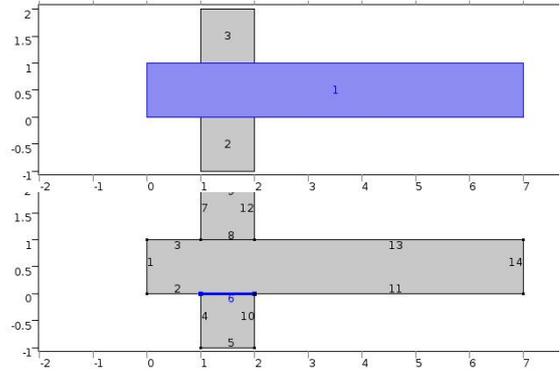


Fig. 3. At the top, the tree main domains at $t = 0$. Region 1 for water, and region 2 and 3 for oil. At the bottom, the interphases are denoted by lines 6 and 8 where fluids are in contact at $t = 0$.

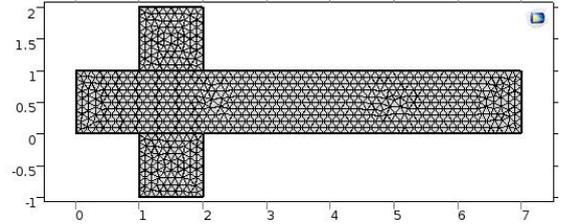


Fig. 4. Coarse mesh in the geometric design.

The mesh will define the size of the elements in which the geometry is divided, and then, the problem should be solved through the Finite Element Method. Finer meshes give better results but require more significant computational resources [30].

We used a coarse mesh (Fig. 4), which gave 1398 domain elements, which represents 9360 degrees of freedom when solving a two-dimensional model. The domain elements were solved for velocity u , pressure p and advection at the interface φ . As this study is time-dependent, we proposed a range from 0 to 0.1 seconds with a step of 0.001 seconds, to ensure the convergence.

All simulations were done in a desk computer with four processors (quad-core) Intel i7 and 32Gb of RAM with Windows Server. Each simulation took between 14 to 24 hours to run and used less than 2Gb of RAM.

IV. RESULTS AND DISCUSSION

In Fig. 6, in blue, we observe water inlet velocities variation from 0.001m/s to 0.009 m/s, with velocity steps of 0.002 m/s, inside vegetable oil carrier fluid at 0.01m/s in both inlets. We observed the relation between velocity, size, and rate of water droplet production. According to Baroud and Utada, there are three regimes possible for $x = width_{in}/width_{out}$ [5, 21]. In the first one, the fluid changes and reaches stable parallel regime flow (jetting), passing through the squeezing regime and droplet regime. In our case $x = 0.5$ so, droplets are produced before they block the channel due to viscous *shear-stress* overcoming interfacial tension. We do not observe squeezing regime since the width of the channel was $x \ll 1$, then droplets do not obstruct the flow of continuous phase.

At $v = 0.02$ m/s of water inlet velocity or higher we found Rayleigh-Plateau instability and the jet develops undulations that lead to dropping formation downstream (Fig. 5b-c).

Fig. 7 shows a pressure profile along the microfluidic channel (1 μm width) at water inlet velocity of $v = 0.005$ m/s, which is useful to trace the changes in pressure inside and outside of droplets. A variation of pressure is observed at the interphase of droplet along the microchannel, which is in agreement with theory [21].

Then, in all cases, droplet production can be achieved until a water inlet velocity limit. We observed the inverse relation between the droplet distance and the inlet velocity. If the distance between droplets decreases, the inlet velocity increases.

However, when water inlet velocity increases to reach the Weber number larger enough to overcome interfacial tension, a continuous flow of water is achieved. This occurs when $v = 0.02$ m/s for water.

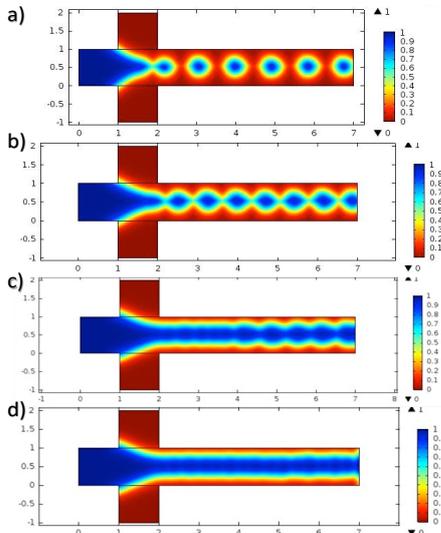


Fig. 5. Microfluidic simulation of corn oil and water (blue) flows at ,20°C, $t = 0.05$ s volume fraction at a) 0.01 b) 0.02, c) 0.05, and d) 0.06 m/s. Formation of instability of water were carried out at inlet velocity of 0.02 m/s or higher.

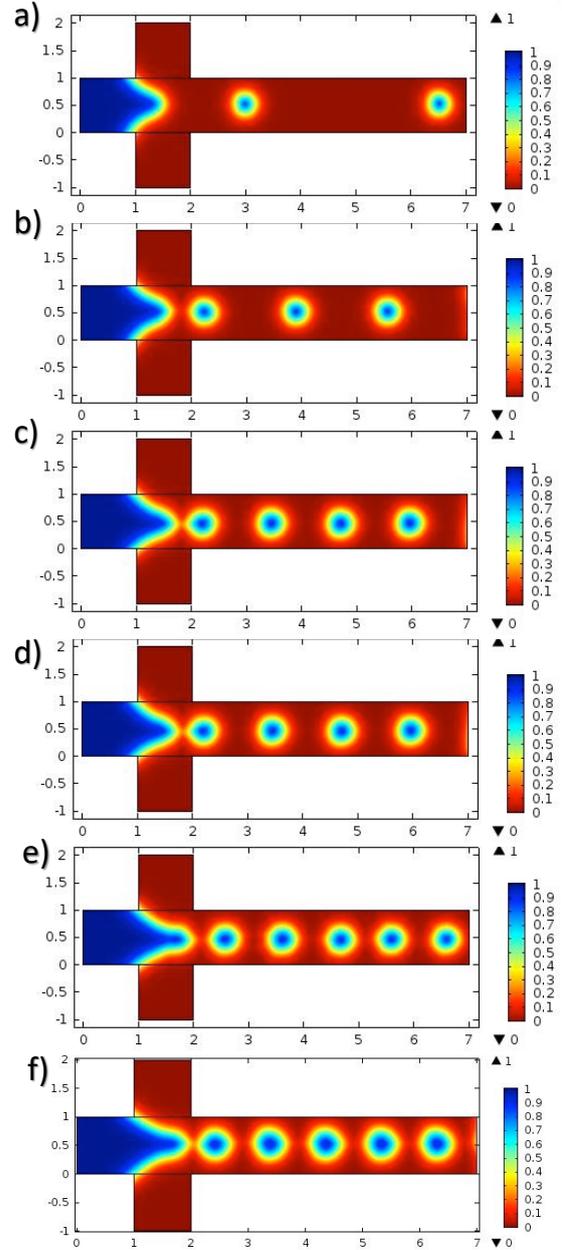


Fig. 6. Microfluidic simulation of corn oil and water (blue) flows at 20°C, $t = 0.05$ s volume fraction at a) 0.01 and b) 0.02.

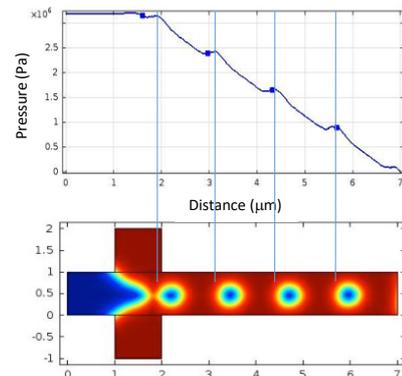


Fig. 7. Pressure profile along the microfluidic channel at water inlet velocity $v = 0.005$ m/s.

V. CONCLUSION

Microfluidics computational simulation was used to establish the conditions, in a time-dependent two-phase microfluidic flow in 2D with a "T" cell geometry with 1 μm of width, for microdroplet fabrication. Microdroplet formation was carried out for inlet water velocity from 0.001 to 0.02 m/s as velocity limit. Over 0.02 m/s, we found instability of water and no oil microdroplet formation. Also, we do not observe squeezing regime since the width of channels where $x \ll 1$, and then droplets do not obstruct the channel as it grows, neither restricts the flow of continuous phase.

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