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INTERACTION OF TWO DROPS FLOWING THROUGH A MICROCAPILLARY

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Abstract. *We study the flow of two drops suspended in a continuous phase through a straight micro capillary. The evolution of the drops interface is determined by a modified level set method which is incorporated to a fully coupled implicit Navier-Stokes solver based finite element method. We investigate the effect of an initial distance between the two drop, as well as the effects of the interfacial tension on the drop speed. The viscosity ratio and flow rate were kept constant in the results presented here. The initial distance between the drops indicates whether the flow of one drop affects the other or if they flow as two distinct single drops. The lower is the capillary number the more difficult is to the drops to interact with each other.*

Keywords: *Emulsions, Level-SetMethod, Train of Drops, Capillary Hydrodynamics, Finite Element Method*

1. INTRODUCTION

Capillary two-phase hydrodynamics have three considerable distinctions from two-phase flows in microsystems. First, there is an increase in the ratio of the surface area of the phases to the volume that they occupy; second, the flow is characterized by small Capillary numbers at which the surface tension forces predominate over viscous forces; and third, the microroughness and wettability of the wall of the microchannel exert a considerable influence on the flow pattern formation. Recent development in micro fluid dynamics can be found in several application such as in microreactor (Kashid and Agar, 2007; Kashid *et al.*, 2005), in processes of liquid-liquid extration (Kashid *et al.*, 2007), in transport of emulsions in porous media, which is relevant in several other applications such as enviromental science (Ouyang *et al.*, 1995; Allan *et al.*, 2006), bioengineering, greenhouse storage, oil recovery, among others (Hofman and Stein, 1991; Tallaskstad *et al.*, 2009; Idowu and Blunt, 2010). Besides, the flow of emulsions can be used as a model for the flow in pore scale.

The basic flow phenomena of emulsion transport through pore throats are well discribed by the flow of a single drop through a constricted microcapillary. The dynamics of immiscible drops flowing through a capillary tube with a diameter that varies sinusoidally 0was fisrt studied experimentaly by Olbricht and Leal (1983). Tsai and Miksis (1994) investigated using a boudanry integral method the creeping flow of a drop suspended in a Newtonian liquid in a constricted capillary, focusing on the drop breakup that occurs at certain operating conditions. The effect of the dispersed phase on the pressure drop was decribed by Cobos *et al.* (2009) showing that the flow rate relation in the flow through capillaries can be represented by a mobility reduction factor, which is defined as the ratio of the pressure drop for the continuous phase flow to the average pressure drop of the emulsion flow, at the same flow rate. Roca and Carvalho (2013) studied the flow of a single drop suspended in a continuous immiscible phase flowing thhrough a constricted micro capillary. The transient free boundary capillary flow problem was solved by coupling a modified level-set method to a fully-implicit finite element method. The results found by Roca and Carvalho (2013) led to a better pore-scale flow model for a wide range of parameters.

However, there are multiple drops in an emulsion that could interact with each other during the flow and this interaction is not well defined. There are several works in the literature on microhydrodynamics. Several studies can ben found in the literature on gas-liquid systems (Schwartz *et al.*, 1986; Ratulowski and Chang, 1989), other studies focus on the ydrodynamics of liquid-liquid systems, with different flow patterns such as slug (Jovanovic *et al.*, 2011; Bico and Quéré, 2000) and drop flow (Danov *et al.*, 2003; Baroud *et al.*, 2010; Westborg and Hassager, 1989). Nevertheless, most of the studies in drop flow focus the attention in the flow of a single drop. In most cases when more drops are considered in the flow their interaction is studied only in cases of coalescence as can be seen in the work of Danov *et al.* (2003) and Baroud *et al.* (2010). In the work of Bico and Quéré (2000) trains of juxtaposed drops in a tube are described and found to move spontaneously, in wetting conditions, beacuse of their asymmetry, but their main focus in on the coating proprieties of the lubricating film left by the first drop.

Here, we solve the transient flow of two drops suspended in an immiscible liquid through a straight micro capillary using a modified level-set method incorporated to a fully coupled implicit Navier-Stokes solver based finite elements method. The method and its numerical implementation were validated by Roca and Carvalho (2013).We focus on the interaction of the two drops in their flow through the capillary. The results for a strapight capillary show the evolution of two drops as they flow through the capillary. The results for a straight capillary show the evolution of the drops

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deformation and the influence of the gap distance between the drops as well as the influence of the interfacial tension in the behavior of the flow of the drops.

2. MATHEMATICAL FORMULATION

2.1 Navier-Stokes equation and boundary conditions

Drops of oil (viscosity μ_o), each with volume $\frac{1}{6}\pi d_{drop}^3$, are suspended in water (viscosity μ_w). We assume that both phases have equal density, $\rho_w = \rho_o = \rho$, such that gravitational forces can be neglected and the flow considered axisymetric.

The liquids flow through a $1.0mm$ long straight cappilary with radius of $D/2 = 50\mu m$ as shown in Fig. 1.

Figure 1: Sketch of the flow domain with two oil drops

The velocity and pressure fields of incompressible, axisymetrical flow of Newtonian immiscible fluids with equal densities are governed by the continuity and momentum equations.

$$\nabla \cdot \mathbf{u} = 0, \tag{1}$$

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right] = \nabla \cdot \mathbf{T}_i + \mathbf{f} \tag{2}$$

where ρ is the density of both phases, $\mathbf{T}_i$ is the stress tensor of each phase i. For Newonian liquids, it is given by $\mathbf{T}_i = -p\mathbf{I} + \mu_i[\nabla \mathbf{u} + (\nabla \mathbf{u})^T]$, where $\mathbf{I}$ is the Identity tensor, p is the pressure, μ_i is the viscosity of each phase and $\mathbf{f}$ ia a body force. In this work, we use $i = w$ for the continuous phase (water) and $i = o$ for the dispersed (oil) phase, as was used by Roca and Carvalho (2013) in their work.

In order to solve the Navier-Stokes system, boundary conditions are needed. The domain is bounded by inflow and outflow planes, a solid wall and a symmetry axis. In addition, two boundary conditions, force balance and velocity continuity given by Eq. (3) and (4), need to be specified along the interlayer that separates the two liquids. The definition of the other boundary conditions are discussed by Roca and Carvalho (2013).

$$\mathbf{n}_I \cdot (\mathbf{T}_w - \mathbf{T}_o) = \sigma \frac{d\mathbf{t}_I}{ds}, \tag{3}$$

$$\mathbf{u}_w = \mathbf{u}_o, \tag{4}$$

where $\mathbf{n}_I$ and $\mathbf{t}_I$ are the unit normal and tangent vectors to the interlayer and s is an arc-length coordinate along the free surface. $\mathbf{u}_i$ and $\mathbf{T}_i$ are the velocity and stress tensor for phase i and σ is the interfacial tension.

A *free boundary problem* is presented by the formulation discussed above. The drop configuration at each time is an unknown and it is a part of the solution.

We use the same approach used by Roca and Carvalho (2013). This approach is based on capturing the interface through the definition of a scalar field called the level-set function. The *level-set method* was fisrt introduced by Osher and Sethian (1988) and is discussed in the next subsection.

2.2 Level-Set formulation

Level-set method was used here to capture the interfaces between the liquids as the drops flows through the capillary. In the level-set approach, a smooch function $c(x)$ is defined over the entire domain such that it has different signs inside the domain region occupied by each of the two different phases, here, $c < 0$, in the oil phase and $c > 0$ in the water phase. The interface is described by the curve at each $c \equiv 0$ (Osher and Sethian, 1988). Tipically, the initial level-set function is defined as the signed-distance $d(x)$ from the interface, defined as:

$$d(x) = min(x - x_i), \forall x_i \in interface. \tag{5}$$

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The evolution of the interface is described by an advection equation

$$\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = 0 \tag{6}$$

In the level-set method, the momentum equations for the two phases can be expressed in an unified way by considering the liquid proprierties at each point of the domain to be a function of the level-set function. The unified viscosity is given by Eq. 7.

$$\mu(\mathbf{x}) = \mu_o + (\mu_w - \mu_o) H_\varepsilon(c(\mathbf{x})) \tag{7}$$

where μ_o and μ_w are viscosities of oil and water repectively and $H_\varepsilon(c)$ is the smoothed Heaviside function, a continuous approximation of the Heaviside function that helps to improve the numerical stability when the viscositu is interpolated across the interface. It is defined as

$$H_\varepsilon(c) = \frac{c + \epsilon}{2\varepsilon} + \frac{1}{2\pi} sin(\frac{\pi c}{\varepsilon}) \tag{8}$$

where ε is a layer along the interface where the Heaviside function is smoothed.

The unified momentum conservation equation for both phases is written as

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right] = \nabla \cdot [-p\mathbf{I} + \mu(c)(\nabla \mathbf{u} + (\nabla \mathbf{u})^T] + \sigma \kappa \nabla c \delta_\varepsilon(c), \tag{9}$$

where κ is the interface curvature, ∇c describes the vector normal to the interface and $\delta_\epsilon(c)$ is a smoothed continuous approximation of the Dirac delta function.

The interface curvature κ is evaluated in terms of the level-set function

$$\kappa = \nabla \cdot \frac{\nabla c}{\|\nabla c\|}. \tag{10}$$

Since there is an inflow plane in the analysis presented here, the value of the level-set function along this boundary needs to be specified at each time step, as a boundary condition. The distance from each point of the inflow boundary to the interface at each time step is not known and therefore the level-set function cannot be defined as the signed distance function. Here we use the smooth function proposed by Roca and Carvalho (2013).

$$c(x) = \begin{cases} 1, & min(\mathbf{x} - \mathbf{x_i}) > \epsilon_c, \forall \mathbf{x_i} \in interface; \\ \frac{d(\mathbf{x})}{\epsilon_c} + \frac{1}{\pi} sin(\frac{\pi d(\mathbf{x})}{\epsilon_c}), & \|min(\mathbf{x} - \mathbf{x_i})\| \leq \epsilon_c, \forall \mathbf{x_i} \in interface; \\ -1 & min(\mathbf{x} - \mathbf{x_i}) < \epsilon_c, \forall \mathbf{x_i} \in interface. \end{cases} \tag{11}$$

It approximates the signed-distance function only close to the interface and has a constant value away from it, as shown in Fig. 2 for a spherical drop placed inside a capillary. Roca and Carvalho (2013) set the parameter $\epsilon_c = 5h$, where h is a characteristic length scale of the finite element mesh.

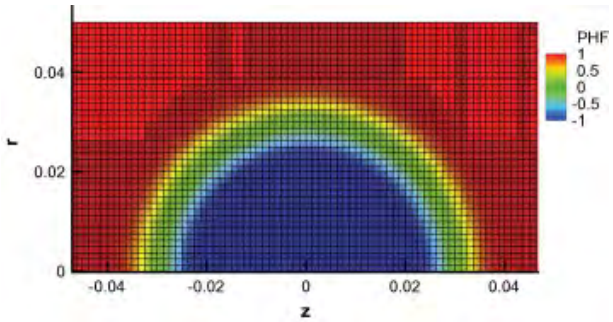

Figure 2: Initial value of the level-set function for a single drop

3. SOLUTION METHOD

Finite element method was used to solve the mommentum and mass conservation equations, Eqs. (9) and (1), coupled with the level-set advection equation Eq.(6). The flow domain was divided into quadrilateral elements.

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The velocity, pressure and level-set function were approximated with a linear combination of a finite number of basis functions.

$$\mathbf{u} = \sum_j \tilde{\mathbf{u}}_j(t)\varphi_j^u = \sum_j [\tilde{u}_j(t), \tilde{v}_j(t)]^T \varphi_j^u, \qquad p = \sum_j \tilde{p}_j(t)\varphi_j^p, \qquad c = \sum_j \tilde{c}_j(t)\varphi_j^c \tag{12}$$

To evaluate the curvature κ at each point of the domain, it is necessary to compute the second derivative of the level-set function. Because the derivatives of the finite elemen basis functions used to approximate the level-set funcion c are discontinuous along the element boundaries, the courvature cannot be evaluated directly. Roca and Carvalho (2013) proposed to use an additional variable $\mathbf{g} = [g_z, g_r]$ to represent the gradient of the level-set function as an independent field wich is also approximated which a linear combination of a finite number of continuous basis functions.

$$\mathbf{g} = \sum_j = \tilde{\mathbf{g}}_j(t)\varphi_j^g = \sum_j [\tilde{g}_{zj}(t), \tilde{g}_{rj}(t)]^T \varphi_j^g \tag{13}$$

Then the courvature κ can be evaluated in terms of $\mathbf{g}$ such as,

$$\kappa = \nabla \cdot \frac{\mathbf{g}}{\|\mathbf{g}\|} = \frac{1}{r}\frac{g_r}{(g_z^2 + g_r^2)^{1/2}} + \frac{g_z^2 \frac{\partial g_r}{\partial r} - 2g_z g_r \frac{\partial g_z}{\partial r} + g_r^2 \frac{\partial g_z}{\partial z}}{(g_z^2 + g_r^2)^{3/2}} \tag{14}$$

The basis functions used to expand the independent variables were: Lagrangian biquadratic polynomials for velocity φ^u and level-set function φ^c, linear discontinuous for pressure φ^p, and Lagrangian bilinear for interpolated level-set function gradient φ^g.

The weak form of the formulaton was obtained by multipluing each equation of the system, Eqs. (12) and (13), by weighting functions, integrating over the physical domain and applying the divergence theorem to the appropriate terms. Galerkin's method was applied to the equations of momentum conservation, continuity and interpolated level-set gradient, i.e the weighting functions are the basis functions. Streamline Petrov-Galerkin weighting functions were used in the level-set equation, $\psi^c = \varphi^c + h_e \frac{\mathbf{u}}{\|\mathbf{u}\|} \cdot \nabla\varphi^c$, where h_e is the upwind parameter.

Essential boundary conditions were imposed by replacing the corresponding weighted residual equation with imposed value of velocity and level-set function. Natural boundary conditions were applied through the boundary integrals that came from the divergence theorem.

Once all the variables are represented in terms of the basis functions, the system of partial differential equations reduces to a set of ordinary differential and algebraic equations that describe the evolution of the coefficients with time:

$$\mathbf{R}(\mathbf{v}, \dot{\mathbf{v}}, \mathbf{f}) = 0, \tag{15}$$

where $\mathbf{R}$ is the set of the weighted residual equations, $\mathbf{v}$ is the vector of basis functions coefficiens, $\dot{\mathbf{v}}$ its time derivative and $\mathbf{f}$ is a vetor of all input parameters (physical properties, geometry of the flow and boudary condition information).

The temporal discretization of the set of equations, Eq. (15), was followed by predictor-corrector algorithm. The predictor step consists of a foward Euler method and the corrector step consists of a first-order fully implicit Euler method, wich is unconditionally stable.

The resulting set of nonlinear algebraic equations at each time was solved simultaneously by Newton's method with an initial estimate resulting from an explicit Euler step from the solution at the previous two time steps, as showed by Roca and Carvalho (2013).

4. RESULTS

The formulation and solution method used here were validated by Roca and Carvalho (2013), comparing the predictions of the flow through a straight capillary with results available in the literature. A mesh with 15x150 quadrangular elements was used to obtain the results presented in this paper.

The discretization of the level-set formulation does not enforce mass conservation. The fully coupled / fully implicit method used to discretize the set of differential equations minimizes this mass conservation problem by choosing the appropriate element size and time step.

As discussed by Roca and Carvalho (2013), the maximum time step to assure mass conservation within a precribed accuracy is a function of the interface velocity, therefore the imposed flow rate, and the element size. They showed that to keep the mass loss smaller than 10% of the original mass of the drop, the time step needs to be smaller then $1/10$ of the traveling time of the drop through a single element.

Figure 3 shows a second drop included in the initial guess of the field of the level-set function with a prescripted gap between the drops.

In the predictions presented here, the viscosity ratio was $\lambda = \mu_o/\mu_w = 10$ and the flow rate was set to $Q = 0.05ml/h$. We studied the evolution of the drops configuration as a function of the capillary number (interfacial tension between both

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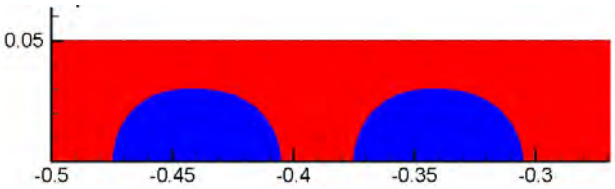

Figure 3: Initial level-set function of a pair of drops $30\mu m$ distant of each other

phases) and initial distance between the drops. For each case studied, the time step was changed until the solution became independent of its value. An example of the time step independency analysis is shown in Fig. 4. The distance between the drops is plotted as a function of time for different values of time steps. The solution becomes independent of the time step for $\Delta t = 3 \times 10^{-4}$. In this example, the initial distance between the drop was $\delta = 30\mu m$ and the interfacial tension effect was neglected, i.e. $Ca = \infty$.

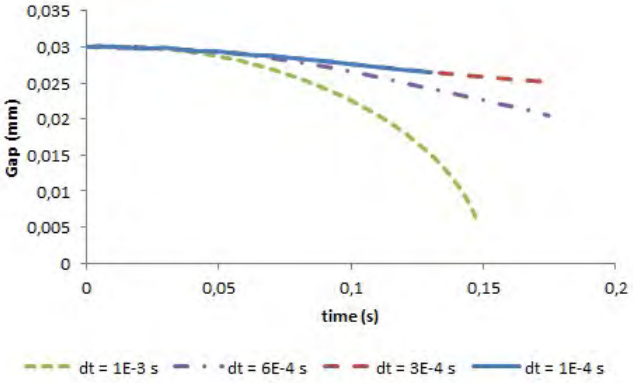

Figure 4: Gap between drops with different values of time step Δt with $Ca = \infty$

Including the effect of the interfacial tension the characteritics of the flow change, therefore it is possible that the time step needed for a solution to become independent of its value could also change. Tests with different capillary numbers ($Ca = 0.75$, $Ca = 0.25$ and $Ca = 0.10$) were made in order to find the time step necessary for each situation. For $Ca = 0.75$ and 0.25 the solution becomes independent for a time step $\Delta t = 3.0 \times 10^{-4}$. For $Ca = 0.10$ the solution did not become independent with the same time step as the other cases. In this case the independecy was achieved with $\Delta t = 1 \times 10^{-4}$.

However, those time steps are chosen to ensure mass conservation. Figure 5 shows the evolution of the mass of the drops over time for different time steps. Although the larger time step is sufficient for the solution to independent, it does not ensure mass conservation. With that in mind, the time step is $\Delta t = 1 \times 10^{-4}$ for all the cases studied.

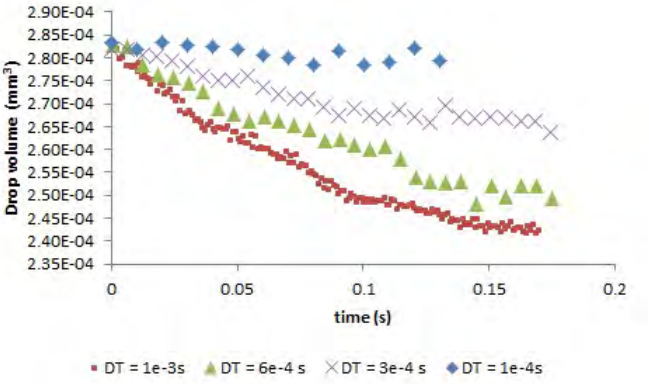

Figure 5: Drop volume for different time steps with $Ca = \infty$

Figure 6 shows the flow of two drops, initially separated by a gap distance $\delta = 30\mu m$, with $Ca = 0.75$ in three different time steps. It can be seen that the distance between them decreases until they start to merge. The coalescence of the drops and it's effect on the flow are not studied in this work.

We will assume that the drop on the left in Fig 6 is drop number one or the fisrt drop, while the one on the right is drop number two or second drop. Knowing the evolution of the position of the extremities of the drops, we were able to

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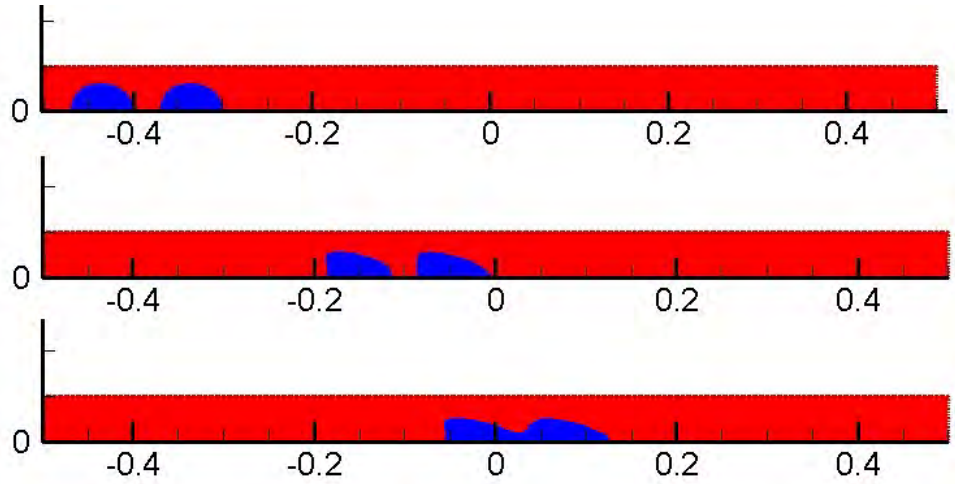
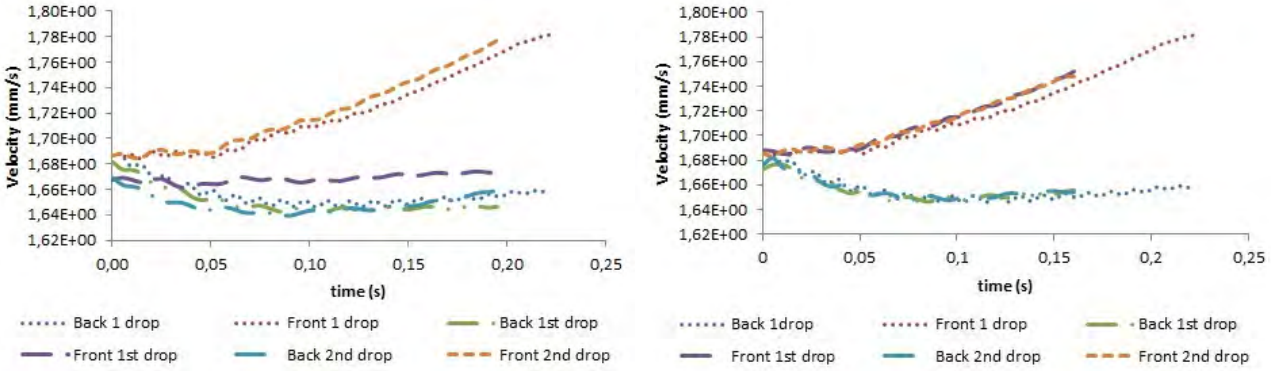


Figure 6: Evolution of the drops with $Ca = 0.75$



(a) $\delta = 30\mu m$ (b) $\delta = 500\mu m$

Figure 7: Velocity of the extremities of the drops at $Ca = 0.75$

calculate by central difference the velocity at those points. Figure 7 shows the evolution of the velocity of the extremities of drops one and two along with the velocity of the extremities of a single drop flowing through the capillary. It can be seen that for a gap of $\delta = 30\mu m$ (Fig 7a) the first drop is influenced by the flow of the second drop. As can be seen in Fig 7a the front of the first drop flows faster then the back of the second drop and the back of a single drop. That's why the gap between the drops decreases. However when the initial gap distance is sufficient big (Fig 7b), both drops flow like a single drop.

In Fig. 8 the size of the two drops with different initial gap distance are compared with a single drop flowing through the capillary. The second drop flows as a single drop regardless the distance to the first drop as can be seen in Fig. 8b. Drop number one does not behave as a single drop flowing through the capillary unless the initial gap distance is big enough. Figure 8a shows that with $\delta < 100\mu m$ the size of the first drop is influenced by the initial gap distance.

The gap distance was normalized with its initial value in order to compare the evolution of the gap between the drops for different initial gap distance. The effect of the capillary number on the behavior of the train of drops is shown in Fig. 9. The interfacial tension forces the drops to become more spherical and changes the way the gap decreases. At lower capillary number the size of the drop decreases due to the bigger interfacial tension (Fig. 9a), hence the gap between the drops increases (Fig. 9b). This means that the drops take more time to close the gap and eventually merge when the capillary number decreases.

5. FINAL REMARKS

A fully coupled / fully implicit modified level-set method together with incompressible Navier-Stokes equation were used to describe the flow of one or more oil drops immersed in water through a straight capillary. The system of differential equations was solved by the finite element method.

A modified definition of the level-set function was used in order to impose the necessary boundary condition along the inflow plane. The fully implicit time integration minimized the flux through the interface inherent to interface capturing approaches, such as the level-set method.

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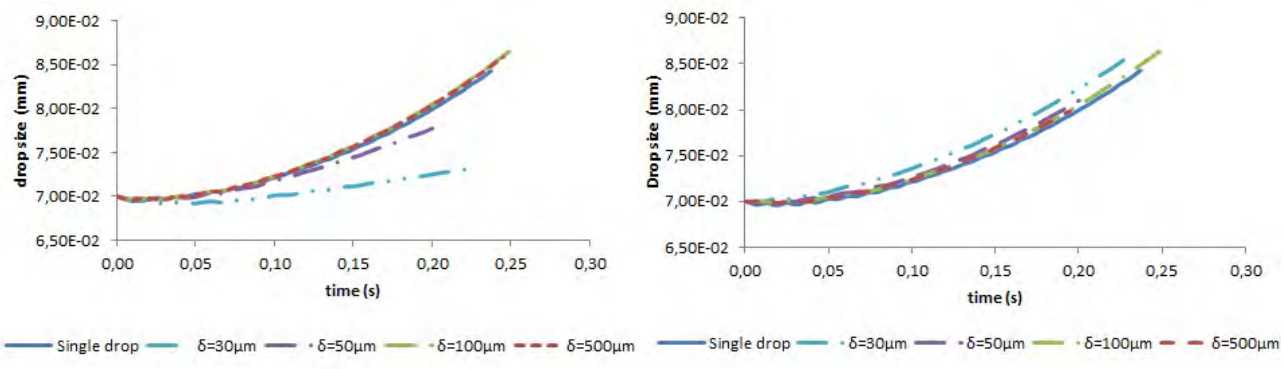
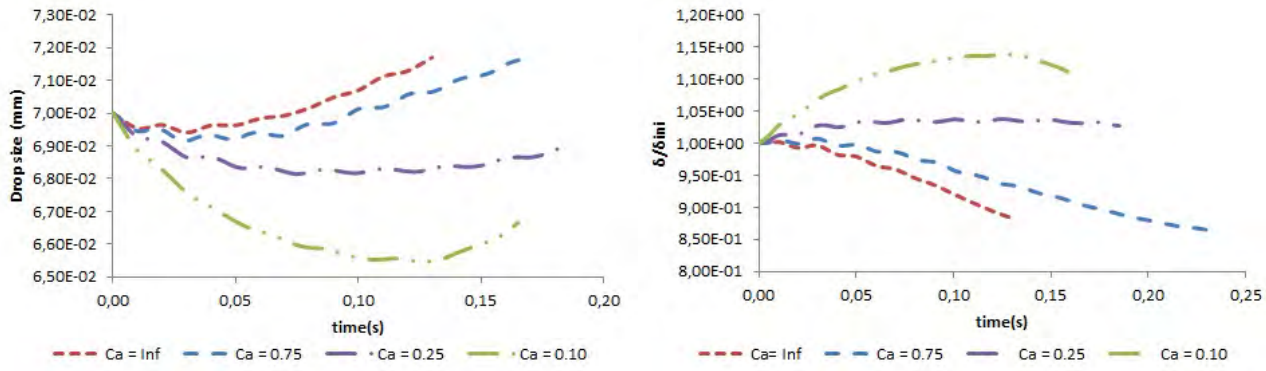


(a) First drop

(b) Second drop

Figure 8: Size of the two drops with $Ca = 0.75$ and different gap distances



(a) Drop size

(b) Gap distance of $30\mu m$

Figure 9: Influence of the capillary number

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The results show how a train of drops flow through a straight capillary. The gap between the drops dictates if the train flows as two distinct drops or if they interact during the flow. This behavior was shown by the influence of the initial gap distance on the velocity of the drops. The interfacial tension creates some dificulties to the flow of the drops, making it more difficult their interaction. All predictions were obtained at a viscosity ratio $\lambda = 10$ and fixed flow rate. The time step used in all simulations was small enough to ensure mass conservation and an independent solution.

6. ACKNOWLEDGEMENTS

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