Electronic Supplementary Material (ESI) for Soft Matter

Supplementary information to the paper "A study of extensional flow induced coalescence in microfluidic geometries with lateral channels"

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1. Visualization of streamlines of the extensional flow using simulation

Streamlines of the extensional flow generated can be nicely visualized using simulation results (see fig.SI1).

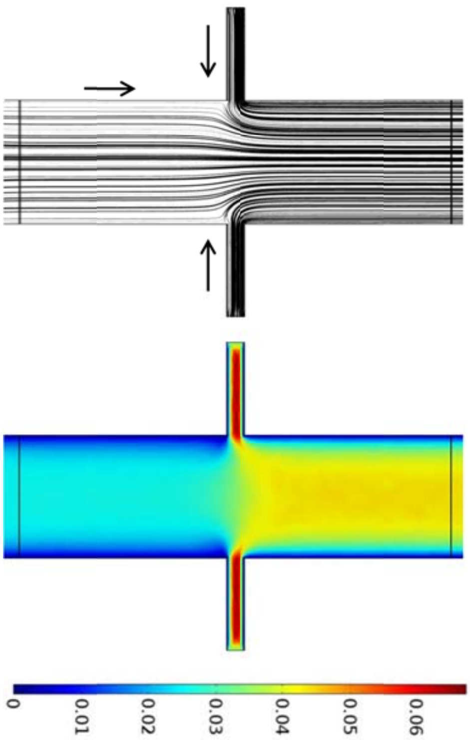

Figure SI1. Simulation results of one part of our microfluidic chip around channel pair C2 (7 mm length). The total flux through the main channel is 0.9 mL/min, while the influx from pair C2 is 0.6 mL/min. Top: plot of a certain number of streamlines. The extensional flow regime can clearly be seen with streamlines converging after the lateral channels. Bottom: Velocity magnitude profile - the legend below shows the scale velocity magnitude, numbers being in m/s.

Clearly, the effects of the drops are completely neglected in the single phase fluid approach, so we also carried out a number of 2D axi-symmetric simulations in order to better understand the interactions between the drops and the continuous phase fluid. Despite this simplification, the results of these simulations brought a good basis of understanding the far field forces acting between drops and determine the range of Reynolds number over the local flow domain. They allowed us to study the evolution of the distance between centers of mass of two droplets advecting along the main microfluidic channel together with the force between these two droplets when passing in front of the lateral control channel. In order to suppress sensitivity with respect to drop deformability, with the concomitant mesh refinement that would result, an artificially large viscosity value was used for the droplet phase. The consequence of this is that the water/oil interface is effectively immobile and the drop capillary number is very small. These conditions for the simulation were chosen because we were interested in the hydrodynamics of the droplet dynamics in the extensional perturbative section independent from system or conditions specificity that would arise from interfacial deformation. However, in experiments, a period of constant separation velocity between centers of mass was observed even for drops of rather high deformability. Consequently, the results of the simulations are of good qualitative value when local features like drop deformation are not focused on (they are of little value in the region close to drop-drop contact).

2. Comment on the need of using a coalescence probability to describe the phenomenon

The profound reasons why the system’s behavior needs to be described using a coalescence probability are not fully understood. Among the perturbations that can impact the coalescence behavior of a certain pair in the chain of drops, the previous event in the chain, as well as the on-going shape relaxation processes, may play a certain role. Nevertheless, the tendency of evolution of coalescence with respect to a given parameter was not affected by such variability, which can be meaningfully averaged over a long observation time (i.e. the behavior, although stochastic, is nonetheless stationary and does not evolve over time).

3. Comment on the use of a film capillary number for describing microfluidic extensional coalescence experiments

The external force applied by use of a couple of lateral channels is intimately coupled to the separation phenomenon itself, and so a fully detailed simulation would be needed to describe the phenomenon for the type of geometry used here, like developed by the group of Leal et al. (see references in the paper). A tentative use of the non-dimensional parameter Ca_f in a simple manner without solving the hydrodynamic problem fully, would actually assume that the stress applied on the lubricating film is constant for the most important part of the separation event. This is highly questionable regarding the geometries used here to separate the first pair of drops which approaches a point source. However the assumption seems likely to be valid when considering coalescence propagation through a chain of drops in which case there is no perturbation flow due to the lateral channels and one might expect that the separating force might remain reasonably constant.

4. Comment on the need of advanced simulation for the description of our results

The actual force pulling apart the drops, and the actual shapes in play, as well as the time profile of the forces, will be different in the case of shape relaxation coalescence and the case of extensional coalescence induced by infusion from a pair of lateral channels. Since there are very subtle effects at play such that we suggest that detailed simulation is required to predict the quantitative behavior for coalescence propagation criteria, e.g. using three drops initially pressed against one another, with one coalescence event occurring at time zero.

5. Comment on the collision offset parameter in experiments by the group of Leal

Note that in the four-roll mill experiments reported by the group of Leal [7,40,41,55], one may think of the collision offset as the variable controlling drainage time, although data with different offset values cannot be compared in a simple manner with results obtained in the present work. In their work, effects of concentration were present neither on coalescence time nor on the critical capillary number for coalescence suppression, and there is absolutely no mention of tip-threading or additional instability development.

6. Comment on the expected influence of the hydrophilic-lipophilic balance of the surfactant on tip-threading, and its relation to extensional coalescence

It can be noted that for PGPR concentration above the cmc, a large body of structural forces due to micelles could strongly contribute to stabilizing the drops against coalescence. Both these mechanisms bring arguments to explain why large surfactant concentrations are used, well beyond the cmc in MCT oil, to stabilize water-in-oil emulsions against coalescence. One can note that in cases known to us, e.g. PGPR in food oil, the oil viscosity is barely affected by the highest surfactant concentration levels used. The same mechanism was observed to be much less likely when using water-soluble surfactant, at least not with small molecular weight surfactants like Tween 20. Hence, it suggests that the influence of surfactant HLB value (hydrophilic-lipophilic balance) is very important in allowing tip-threading. This is consistent with the observation that satellite drops form more easily when using an oil-soluble surfactant like PGPR compared to when using a water-soluble one such as Tween 20. Note that all these comparisons cannot be extrapolated to systems where the surfactant could impart a significant (even local) interfacial viscosity.

7. Videos of experiments

The main flow on the videos is from the right to the left (whereas it was chosen to display all manuscript figures with a flow from the left to the right).

7.1. Video of experiments illustrating figure 3 (name : video_fig3)

Video of extensional coalescence behavior taken in the exact conditions corresponding to figure 3 (both for geometry and system). System : oil phase : PDMS 99 mPa.s. Water phase : water + 0.1% Tween 20. Q_{oil} = 0.12 mL/min. Q_{water} = 0.04 mL/min. Q_{infuse} = 0.12 mL/min. Lateral infusion channels were the pair C1. The total video duration is nearly 8 seconds.

7.2. Video of experiments illustrating figure 3 (name : video_fig5)

Video of extensional coalescence behavior taken in the exact conditions corresponding to figure 5, top part (both for geometry and system). One can observe an avalanche of coalescence events in a chain of drops with the development of a Rayleigh instability eventually yielding break-up into several drops. The total video duration is nearly 10 seconds.