# Flow visualization of a forming droplet in a micro channel with FlexPIV

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## ABSTRACT

This work investigates monodisperse water droplet generation into sunflower oil (SFO) using coflow microchannels with only food-grade solutions of Tween-surfactants and hydroxylpropylether guar gum (HPGG) as disperse phase. We were able to map simultaneously (inner and outer flow behaviour) the entire flow situation given by the geometries, process conditions, and material parameters, utilizing streak imaging (StrIm) and microPIV ( $\mu$ PIV) including the new FlexPIV software and the additional FlexGrid module.

## **1. INTRODUCTION**

Monodisperse emulsions are often required to produce wellstructured multiphase, as well as for lab-on-the-chip applications in food, pharmaceutical, personal care, and cosmetic industries. The drop size and size distribution, interfacial properties, and structuring elements in emulsions determine quality characteristics of the final product. Monodisperse emulsions are also used in fundamental droplet studies as well as a precursor for encapsulation studies because the interpretation of experimental results is much simpler [1-2].

Narrow droplet size distribution is generally difficult to achieve even though modern dispersing devices (e.g. membrane emulsification, pressure homogenizers, rotor-stator toothed disk turbines, opposed jet micro fluidizers, etc.) have been developed in order to produce almost monodisperse emulsions. In most of these techniques the flow field and the stresses acting on the single droplets are not well defined [3]. Microchannel techniques, however, provide a simple and great tool which allows the downsizing of industrial devices utilizing the scaling effects. Numerous benefits such as reduced sample amount, high sensitivity, short analysis time, and parallel processing lead to a simpler investigation of complex processes [4-6]. Studies of micro droplet creation in microchannels formed in different geometries (mostly Tchannel or flow-focusing) have been the subject of many research studies using pure, well characterized solutions and do not take into account the behaviour and interaction of foodgrade and natural products [7-9].

The coflow dispersing technique which we employed within this work, utilizes the direct injection of a disperse phase into an ambient, continuous phase via capillary [10]. We focused on single water in oil droplet formation (w/o) using only foodgrade Tween-surfactants, polymer, and oil solutions for both the dispersed (droplet) and continuous (bulk) phase. The objective was to visualize the influence of single process and material parameters on the droplet dynamics, as we mainly varied the flow rate of both phases and the interfacial and elastic properties of the droplet solution [11]. However, to understand and analyse drop formation and breakup dynamics and also in order to design micro-flow devices and droplet based reactors, it is important to measure the dispersed and continuous flow fields in and around a droplet.

## 2. MATERIAL

#### 2.1 Continuous phase

Commercially available, cold-pressed sunflower oil (SFO, Florin AG) gained from sunflower seeds, was taken as the ambient bulk phase for all experiments. This oil is widely used in nutrition and food industry especially for uncooked foods (e.g. salad dressings, margarine, mayonnaise) because of its high content of vitamin E and unsaturated fatty acids. Even though it is natural product it shows rather constant zero shear viscosity at 56 mPa·s over a range of shear rates.



**Figure 1**.  $\mu$ PIV setup using: (A1) Nd:YAG-double pulsed laser, (A2) mercury lamp, (B) epi-fluorescent prism, (C) microscope objective lens, (D) microscope stage, (E) micro device, (G) barrier filter (transferred  $\lambda$  > 590 nm), (H) CCD SensiCam QE kit, (F) Harvard Syringe pumps.

## 2.2 Disperse phase

For the hydrophilic droplet phase we used non-ionic Tween 20, 40, 60, or 80 surfactants added to distilled water at different concentrated from 1 cmc (beginning of surfactant micelle formation) up to 1370 cmc ( $\sim 2$  V%; most surfactants are in form of micelles). The density and the zero shear viscosity of

the Tween-water mixtures were measured to be water-like. Tween surfactants at higher concentrations are generally applied in food and cosmetic industry to produce and stabilize various kinds of emulsions.

In order to study the extensional influence of the disperse phase on the droplet kinetics and final droplet size, we added hydrophobically modified guar gum (HPGG; hydroxylpropylether guar gum, also called JAGUAR) to the water phase (with 0.01 to 0.2 wt%) [12].

## 2.3 Particle seeding

Different sized fluorescent (nile red) polymer microspheres (carboxylated polystyrene beads) as tracer particles (Duke Scientific) were used to seed both the disperse ( $D_{R700} = 0.7 \mu m$ ) as well as the continuous phase ( $D_{R300} = 0.3 \mu m$ ). The particles show a maximum excitation wavelength of  $\lambda_{ex,max} = 542 \text{ nm}$  (green) and an emission maximum wavelength of  $\lambda_{em,max} = 612 \text{ nm}$  (orange-red). The right choice of seeding density (droplet phase 0.03 V% max., continuous phase 0.01 V% max.) was important allowing us to catch the in-focus particle over the background glow.

## **3. EXPERIMENTAL SETUP**

In order to investigate the flow stream behaviour, we performed  $\mu$ PIV and StrIm experiments employing PDMS coflow microchannel with different capillary geometries. The soft-lithography technique to manufacture PDMS-channels utilized in the present work is a common expertise which is already described elsewhere [9, 13].

Figure 1 illustrates and explains the experimental setup utilizing an inverted microscope (Leica), a PC workstation including Dantec Dynamics PIV/LIV capturing and analysing system.

## 4. ANALYSIS METHOD

#### 4.1 FlexPIV software

Since the development of PIV (particle image velocimetry) more than 20 years ago, many extensions and enhancements to the technique were accomplished. Starting from autocorrelation, going to cross-correlation and further to adaptive cross-correlation (nowadays the de-facto standard in PIV processing) the power of PIV increased tremendously. Methods for special applications, like average correlation for uPIV or hybrid techniques combining PIV and PTV (particle tracking velocimetry) for increased spatial resolution, found their way into the community. The last leap forward was denoted by the consideration of not only translational shifts but also of strain and rotational deformations of the particle pattern ("deformed Windows"). The results of all these methods, however, are velocity fields within a Cartesian (typically square) frame, in which all vectors are calculated using the same constant parameters (e.g. size of the interrogation area (IA), validation setup, etc) within an image. An individual consideration of the local geometry and flow situation does not take place. Only the development of a method for flexible, flow and geometry adapted PIV processing (FlexPIV) remedies the limitations mentioned before.

Using FlexPIV, a concept already known from Computational Fluid Dynamics (CFD) is transferred to PIV applications. In CFD it is commonplace long since that the computational grids are adapted to the nature of the flow and the flow region geometry. This approach yields advantages for PIV methods such as:

- Velocity vectors will only be calculated in defined areas (objects) where a grid has to be set before. Although it is possible to exclude certain areas from being processed by masking out regions of the particle images, the total image is still processed by the cross-correlation algorithm.
- The number and position of the velocity vectors are decoupled from the size of the interrogation areas.
- Grids of different density can be generated adapted specifically to the conditions of the flow field at certain positions. Additionally, it is possible to define different processing parameters for each grid independently.
- As wall structures or boundary lines cross the defined interrogation areas at oblique angles, errors in the calculated velocity vectors may occur. The possible definition of a "wall object" prohibits false calculations by disregarding the part of the interrogation area that belongs to the wall structure.

#### 4.2 Grid generation with FlexGrid module

Prior the velocity flow field calculations a grid similar to CFD applications needs to be defined for each grid-object using a software module FlexGrid. FlexPIV analysis relies finally on the grid definitions provided by FlexGrid. However, the new module offers various object geometries (rectangles, squares, ellipses, circles, and polygon curves) which can be drawn graphically using mouse commands or by direct numerical input. Each object can then be connected to a specific grid type such as Cartesian, polar, elliptic or Delaunay grid (**Figure 2a-d**). The final processing parameters for the cross-correlation output are designated to each grid individually.



**Figure 2**. Illustration of grid-objects (rectangle or square, circle, ellipse, polygon) and grid types (Cartesian, polar, elliptic, and Delaunay grid).

Furthermore, the software module allows providing the defined grid-object with one of the additional properties such as "grid", "hole", "false hole" or "wall". Using an object with the property "hole" for example, areas within another grid can be excluded from the processing (**Figure 2c**). An object with the property "false whole" is used to define a grid within a grid without calculating redundant vectors in the common area. The object property "wall" does not take into account the grey level values covered by this object during cross-correlation. A complex grid definition and the resulting velocity field are shown in **Figure 3** exemplified in a cylinder wake.

Applying FlexPIV including FlexGrid instead of conventional rigid grids, it is feasible to process PIV images in a flexible way which is adapted to the actual flow situation. Hence, the achieved results with FlexPIV exceed what has been known so far. In this work we will present an example of flow analysis, where we visualize simultaneously the inner and outer flow of

a forming droplet in a microchannel utilizing  $\ensuremath{\mathsf{FlexPIV}}$  and  $\ensuremath{\mathsf{FlexGrid}}.$ 



**Figure 3**. Example of grid-object and grid type definitions using the FlexGrid module (A) and the related velocity profile calculated with FlexPIV (B).

#### 4.1 FlexPIV analysis

We performed FlexPIV analysis utilizing an adaptive correlation algorithm with IA-overlap of at least 25-50%. The IA were sized either 16x16 pxl or 32x32 pxl depending on the velocity flow fields, so there was only little velocity variation (< 5-10%) within a IA. Further post-processing procedures were kept to a minimum in all analysis of the presented  $\mu$ PIV experiments. The area outside the actual microchannel was masked out prior FlexPIV analysis to minimize the number of false velocity vectors. In order to verify the result of the cross-correlation, the original fluorescent micrograph and the corresponding velocity vector field was matched as it is shown in **Figure 4** [11].

#### **3. RESULTS**

Generally, the droplet formation during dripping in coflow systems can be divided in 3 different stages: (i) droplet filling where the droplet is attached to the capillary outlet and expands in all direction till the equilibrium of the net forces is lost (interfacial tension versus detaching forces), (ii) droplet separation starts by forming a neck, and (iii) the neck thins rapidly till the droplet finally pinches off (drag and inertial overcome the interfacial tension force) [10].

**Figure 5** shows the flow stream around a forming droplet at the capillary tip. Applying StrIm, we were able to visualize little vortices using only water or Tween-water solution as the droplet phase. Main differences in the flow stream could only be observed once we added little amount (0.01wt%) of HPGG

polymer to the water. As the extensional properties of the polymer elongates the droplet necking, the flow of the bulk phase becomes smoother and the vortices do not exist anymore [14].



**Figure 4.** Fluorescent micrograph matched with the corresponding vector field of a forming droplet at the capillary tip within a coflow microchannel.

The final Figure 6 presents a water droplet within a microchannel at the stage (i) of droplet filling (compare Figure 5 iii). The experiment was run at flow rates of  $Q_d = 0.1$  ml/h ( $v_d$ = 0.83 m/s, disperse phase) and  $Q_c = 0.5$  ml/h ( $v_c = 0.17$  m/s, continuous phase). The fluorescent micrograph including the frame of the high-speed captured picture shows the actual position of the droplet inside the microchannel. We clearly observe, shown by the different colours of the velocity vectors (Figure 5 middle) that the speed of the disperse phase is the fastest close to the capillary outlet and decreases with increasing distance to the orifice. To analyse the flow profile quantitatively the velocity values of the disperse phase were then taken at 4 different stream-wise positions (constant x-value, parallel to the y-axis). We observed that the symmetrical velocity distribution at varies locations shows a decrease with growing droplet and with increasing distance from the capillary tip.



**Figure 5**. Streak images of a forming  $H_2O$  droplet ( $Q_d = 0.01 \text{ ml/h}$ ) into SFO ( $Q_c = 0.1 \text{ ml/h}$ ) inside a microchannel. Both phases are seeded with particles.

#### 9. CONCLUSIONS

We visualized the (w/o) droplet formation and pinch-off at the capillary tip in coflow microchannels systematically using various food-grade solutions. Differences in the droplet evolution process were observed with the addition of food-grade Tweensurfactants and modified guar (HPGG) to the disperse phase which includes modified droplet sizes, elongated drop creation, and the generation of a thread while pinching off. The latter effect is a result of the elastic stresses developed within the fluid during the dispersing process.



**Figure 6.** Illustration of the inner flow behaviour of a forming droplet at the stage of filling: velocity profile (middle), velocity graph (bottom), corresponding micrographic (top) [14-15].

Due to the drop forming at the capillary tip and the unsteady flow behaviour within our experiments, it was impossible to apply a simple averaging or cross-correlation analysis method. Both analytical methods would only lead to an instantaneous velocity measurement with zero-averaging of random noise peaks. FlexPIV algorithm with the additional FlexGrid module instead, allowed detailed analysis with multiple processing schemes of specifically defined, flexible object grids.

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#### REFERENCES

- Whitesides G.M. (2006) The origins and the future of microfluidics. *Nature*, 442(7101), 368-373.
- [2] Windhab E.J., Dressler M., Feigl K., Fischer P., Megias-Alguacil D. (2005) Emulsion processing - from single-drop deformation to design of complex processes and products. *Chemical Engineering Science*, **60(8-9)**, 2101-2113.
- [3] Joscelyne S.M. and Tragardh G. (2000) Membrane emulsification - a literature review. *Journal of Membrane Science*, 169(1), 107-117.
- [4] Hong J.W. and Quake S.R. (2003) Integrated nanoliter systems. *Nature Biotechnology*, 21(10), 1179-1183.
- [5] Qin D., Xia Y., Rogers J.A., Jackman R.J., Zhao X.M., Whitesides G.M. (1998) Microfabrication, microstructures, and microsystems in *Topics in Current Chemistry*, Springer-Verlag Berlin, Vol. 194, 1-20.
- [6] Hardt S. and Schönfeld F. (2007) Microfluidic Technologies for Miniaturized Analysis Systems, Springer-Verlag New York.
- [7] Anna S.L., Bontoux N., Stone H.A. (2003) Formation of dispersions using "flow focusing" in microchannels. *Applied Physics Letter*, 82(3), 364-366.
- [8] Husny J. and Cooper-White J.J. (2006) The effect of elasticity on drop creation in T-shaped microchannels. *Journal of Non-Newton Fluid Mechanics*, 137(1-3), 121-136.
- [9] Whitesides G.M. and Stroock A.D. (2001) Flexible methods for microfluidics. *Physics Today*, 54(6), 42-48.
- [10] Cramer C., Fischer P., Windhab J.E. (2004) Drop formation in a co-flowing ambient fluid. *Chemical Engineering Science*, 59(15) 3045-3058.
- [11] Duxenneuner M.R. (2009) Visualization, design, and scaling of drop generation in coflow processes, *PhD-Thesis* (No.18277; http://e-collection.ethbib.ethz.ch), Department of Food Science and Nutrition, ETH Zürich.
- [12] Duxenneuner M.R., Fischer P., Windhab E.J., Cooper-White J.J. (2008) Extensional properties of hydroxypropyl ether guar gum solutions. *Biomacromolecules*, 9(11), 2989-2996.
- [13] McDonald J.C., Duffy D.C., Anderson J.R., Chiu D.T., Wu H.K., Schueller O.J.A., Whitesides G.M. (2000) Fabrication of microfluidic systems in poly(dimethylsiloxane). *Electrophoresis*, 21(1), 27-40.
- [14] Duxenneuner M.R., Fischer P., Cooper-White J.J., Windhab E.J. (2009) Simultanous inner and outer flow visualization of generated droplets in microchannel coflow. *Lab on a Chip*, (in preparation).
- [15] Duxenneuner M.R., Fischer P., Windhab E.J., Cooper-White J.J. (2009) Investigation of monodisperse droplet generation in coflow. In. Editor Fischer P., *Proc. 5<sup>th</sup> International Symposium* on Food Rheology and Structure, ETH Zürich Switzerland, 15.-18. June.