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Predict and compare the formation of segmented flow in microsystems: Interest for radiochemical liquid-liquid extraction

A. Vansteene, J-P. Jasmin, G. Vera de La Cruz, R. Brennetot, S. Cavadias, C. Mariet, G. Cote

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Vansteene A.¹, Jasmin J.P.¹, Vera de la Cruz G.², Brennetot R.¹, Cavadias S.², Mariet C.¹, Cote G.³

axel.vansteene@cea.fr; jean-philippe.jasmin@cea.fr

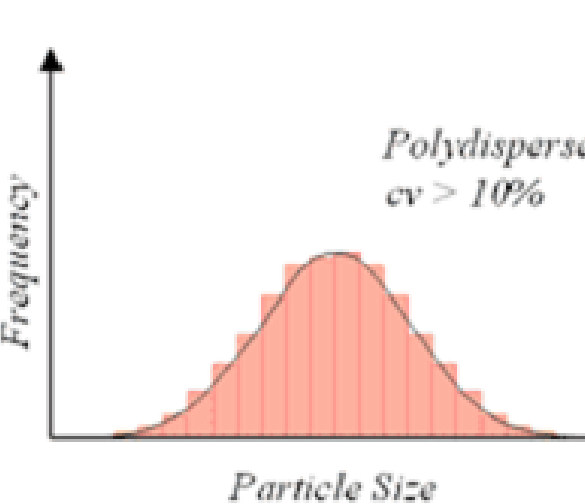
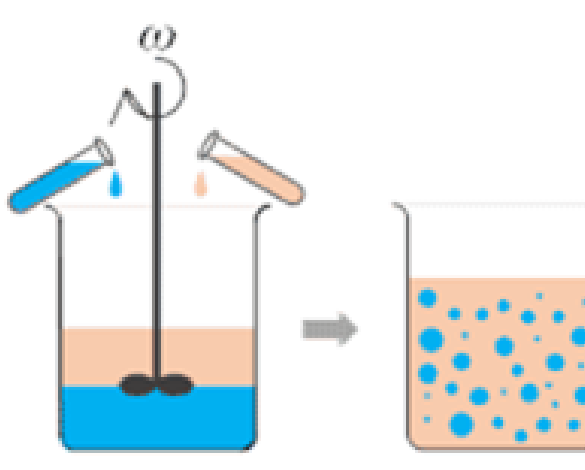
1) Den - Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, F-91191, Gif sur Yvette, France

2) UPMC – Univ Paris 06, 4 Place Jussieu, 75005 Paris, France

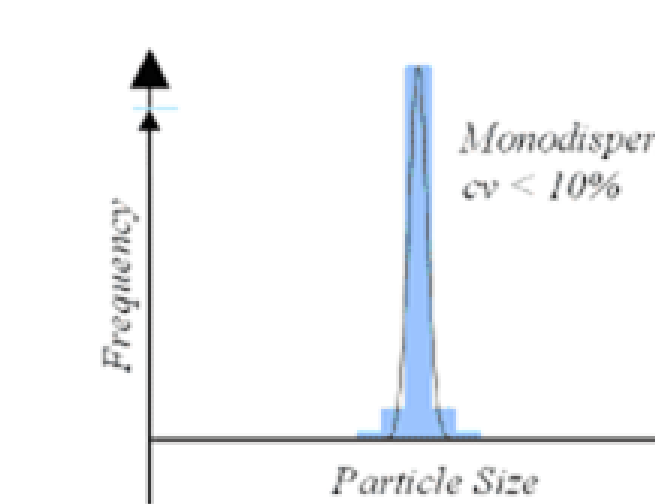
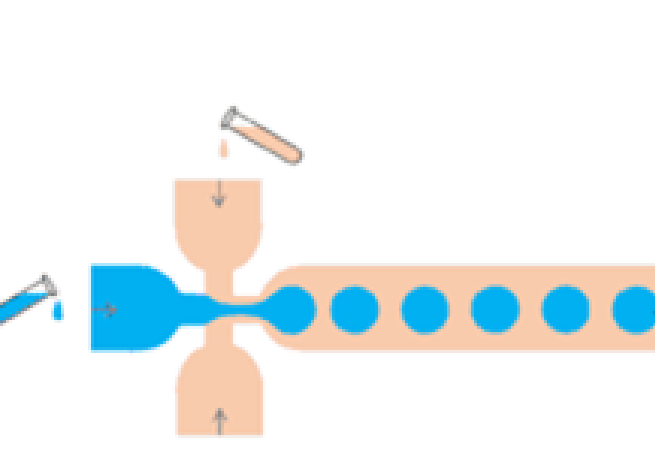
3) PSL, Chimie ParisTech - CNRS, Institut de Recherche de Chimie Paris, 75005, Paris, France

One of the most important separation techniques in radiochemical procedures is solvent extraction. In the last decade, a growing interest in its use in microsystems has emerged because such systems allow a good control of both the interface area between aqueous and organic phases, and the contact time of the two phases. To improve mass transfer in liquid-liquid extraction reactions for radiochemical separation relevant to the nuclear field, our goal is to implement liquid-liquid extraction with segmented flows in microsystems in order to increase the interfacial area. The optimization of the interfacial area requires taking into account the hydrodynamics of biphasic systems. This study is dedicated to the understanding of segmented flows in a T-junction with the aim to form tailor-made droplets for given organic and aqueous phases.

Batch method emulsion production



Microfluidic continuous flow method



ASSETS OF SEGMENTED FLOWS IN MICROSYSTEMS

Microscale assets^[1]

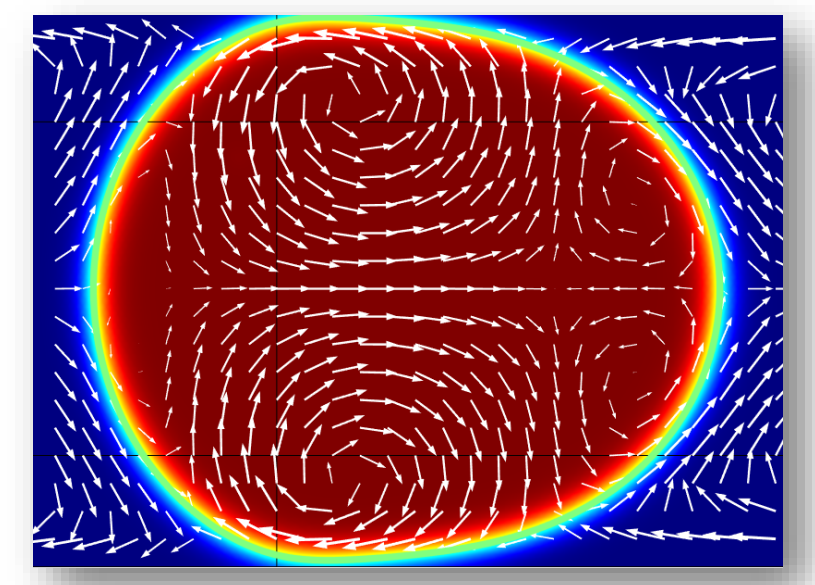
- Volume reduction (from mL to μL)
- Lower operators' exposure to
 - chemicals
 - radiations
- Shorter analysis time
- Lower polydispersity
- Cost reduction



[1] Helle G et al. Talanta, 2015, 139: 123-131.

Segmented flows assets

- Enhanced mass transfer
 - diffusion (by using droplets with a small spacing in between)
 - convection (by recirculating species inside droplets)
- Enhanced interfacial area



■ Organic phase
■ Aqueous phase

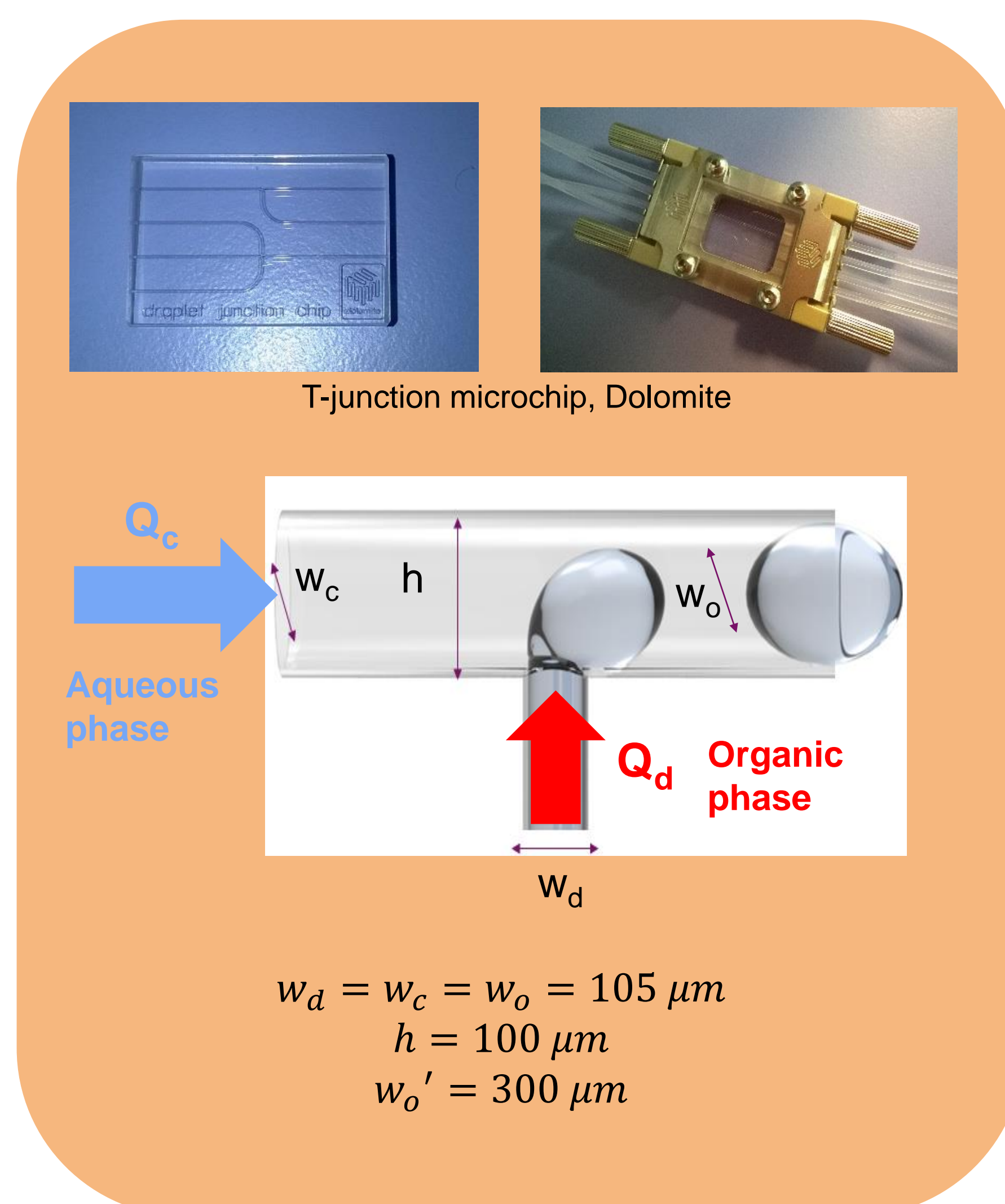
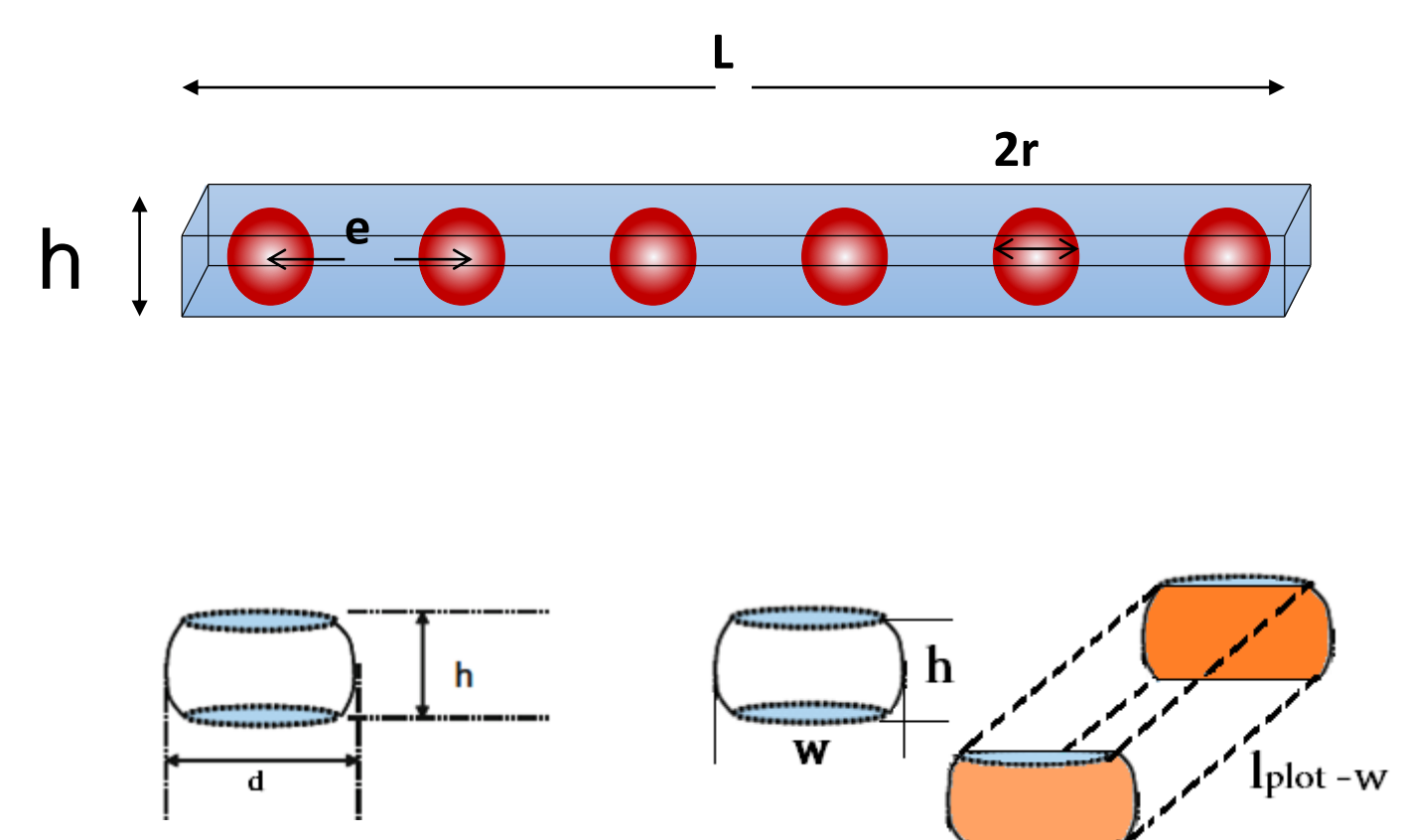
2D CFD Model developed by our team showing evidence of species recirculation inside 70:30 (v/v) dodecane/TBP droplets, in a continuous phase of $[\text{HNO}_3] = 3 \text{ M}$ $[\text{U(VI)}] = 10^{-4} \text{ M}$, with a pressure gradient of $0.66 \text{ Pa}\cdot\mu\text{m}^{-1}$.

HYDRODYNAMICS OF SEGMENTED FLOWS IN A T-JUNCTION

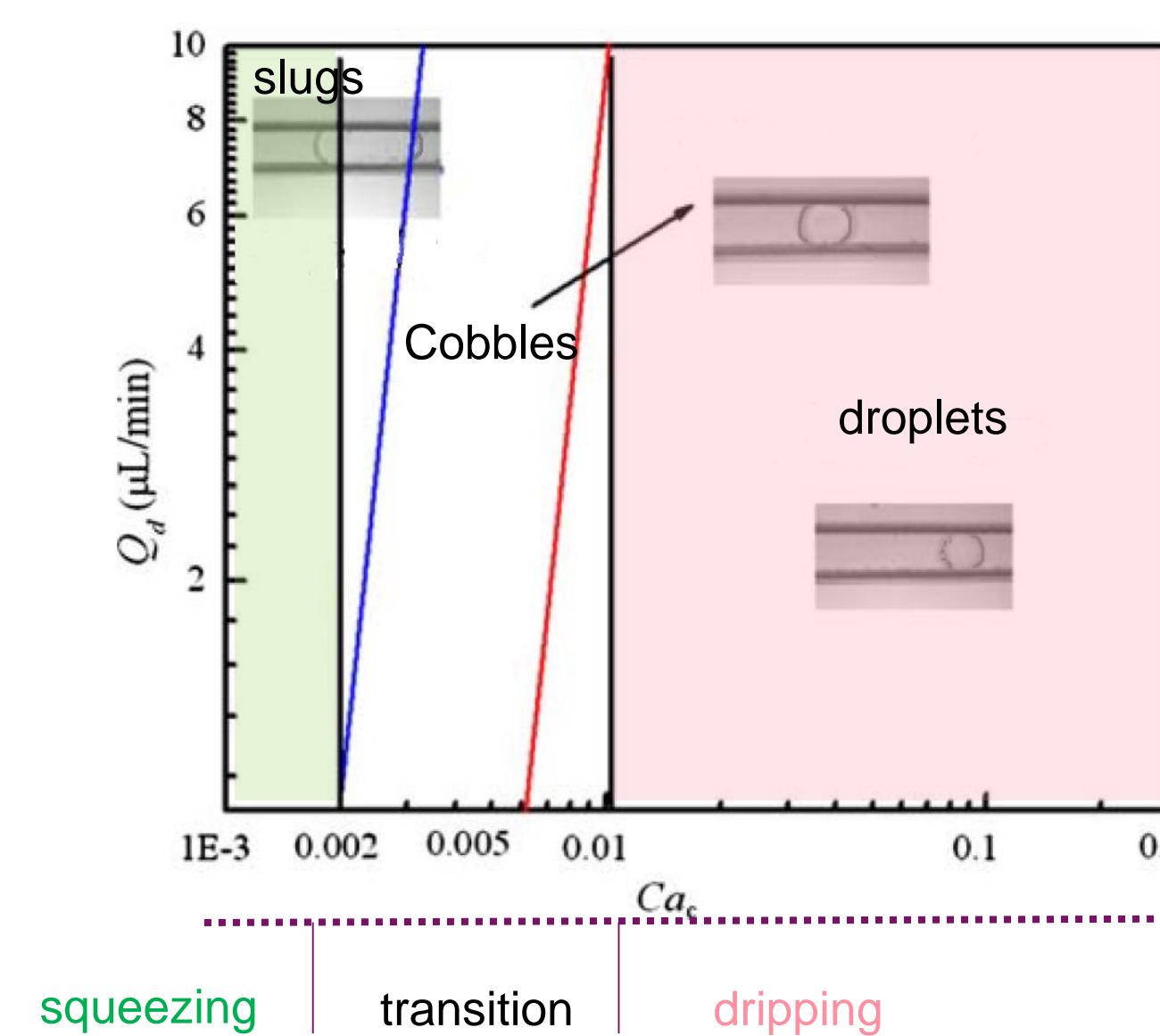
To form tailor-made droplets for given organic and aqueous phases, we identified the geometrical parameters of the T-Junction and the physico-chemical parameters of the aqueous and organic phases involved in the control of the flow regimes in addition to the flow rates.

Segmented flows characteristics

- Droplet volume V_{plug} or length l_{plug}
- Droplet spacing $e = \frac{Q_c + Q_d}{hwf}$
- Generation frequency $f = \frac{Q_d}{V_{plug}}$
- Specific interfacial area $A = \frac{S_{plug}}{ehw}$



Hydrodynamic regimes



Capillary number $Ca_c = \frac{\eta_c Q_c}{\sigma w_c h}$

with

η_c : viscosity of the continuous phase

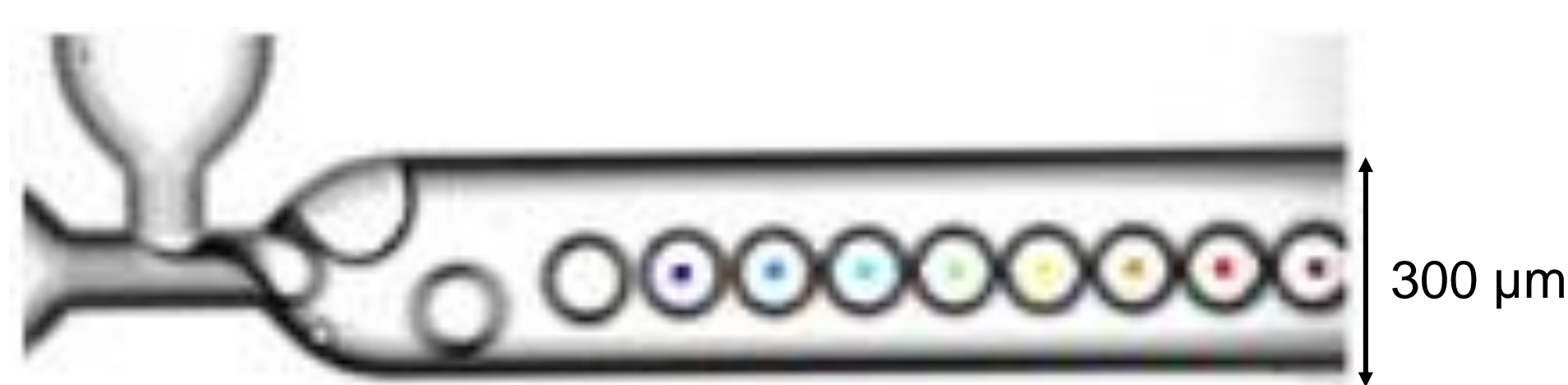
σ : interfacial tension between the aqueous and organic phases

Squeezing regime ($Ca_c < 0.002$)	Transition regime ($0.002 < Ca_c < 0.01$)
$V_{plug} = V_{fill} + \alpha \frac{Q_d}{Q_c} hw_c^2$ [2]	$\frac{l_{plug}}{w_c} = 0.75 \left(\frac{Q_d}{Q_c}\right)^{\frac{1}{3}} Ca_c^{-\frac{1}{5}}$ [3]
[2] Van Steijn, V. et al. Lab Chip, 2010, 10(19), 2513-2518.	[3] Xu, J. H. et al. Microfluidics and Nanofluidics, 2008, 5(6), 711-717.

For the different regimes, we found equations linking flow rates and physico-chemical parameters of the biphasic chemical system with characteristics of the segmented flow (droplet volume, spacing, ...). Therefore, we are able to predict the characteristics of the generated flow.

MODEL VALIDATION FOR H₂O/DODECANE CHEMICAL SYSTEM WITH DROPLET MORPHOMETRY AND VELOCIMETRY (DMV) ANALYSIS

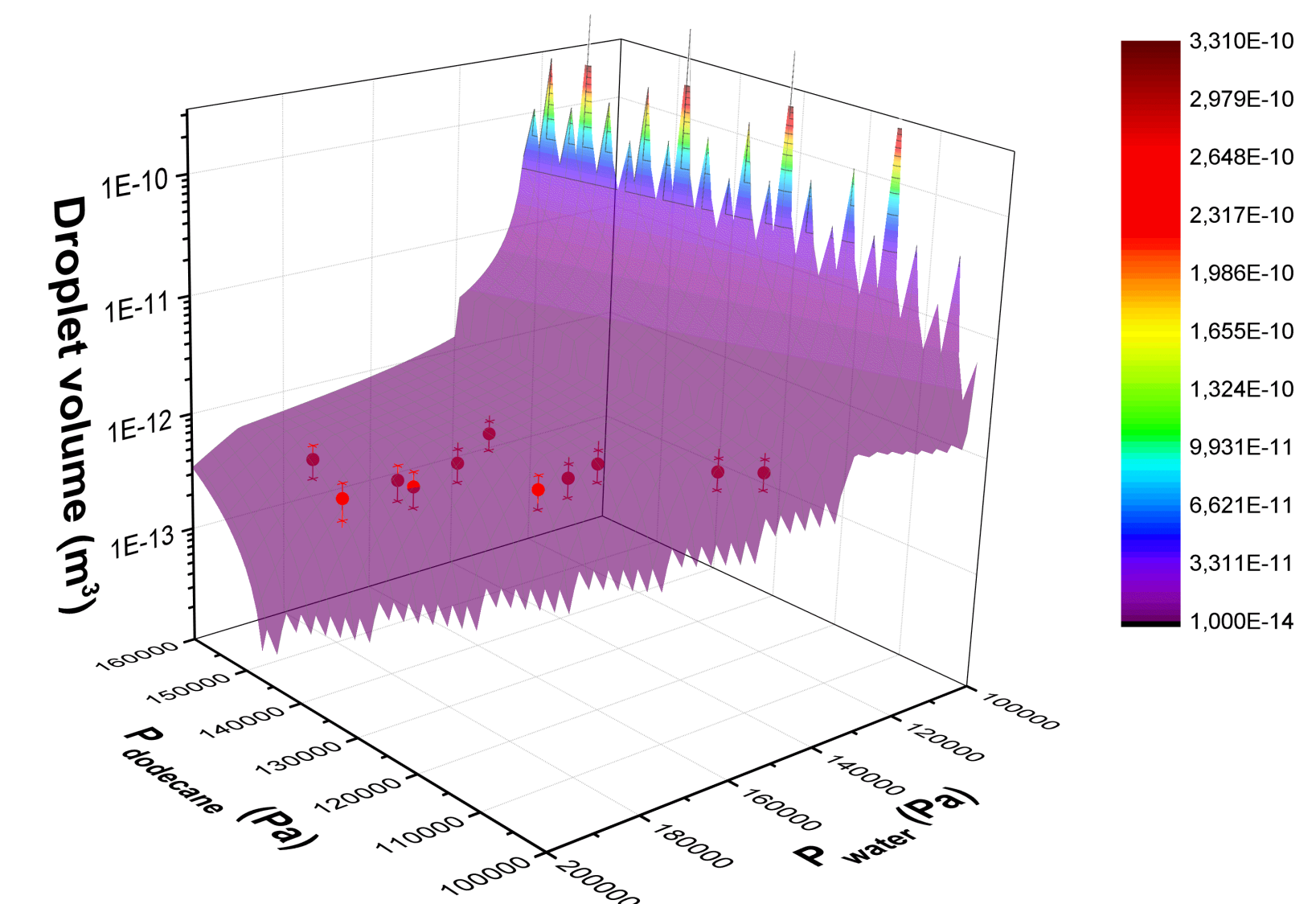
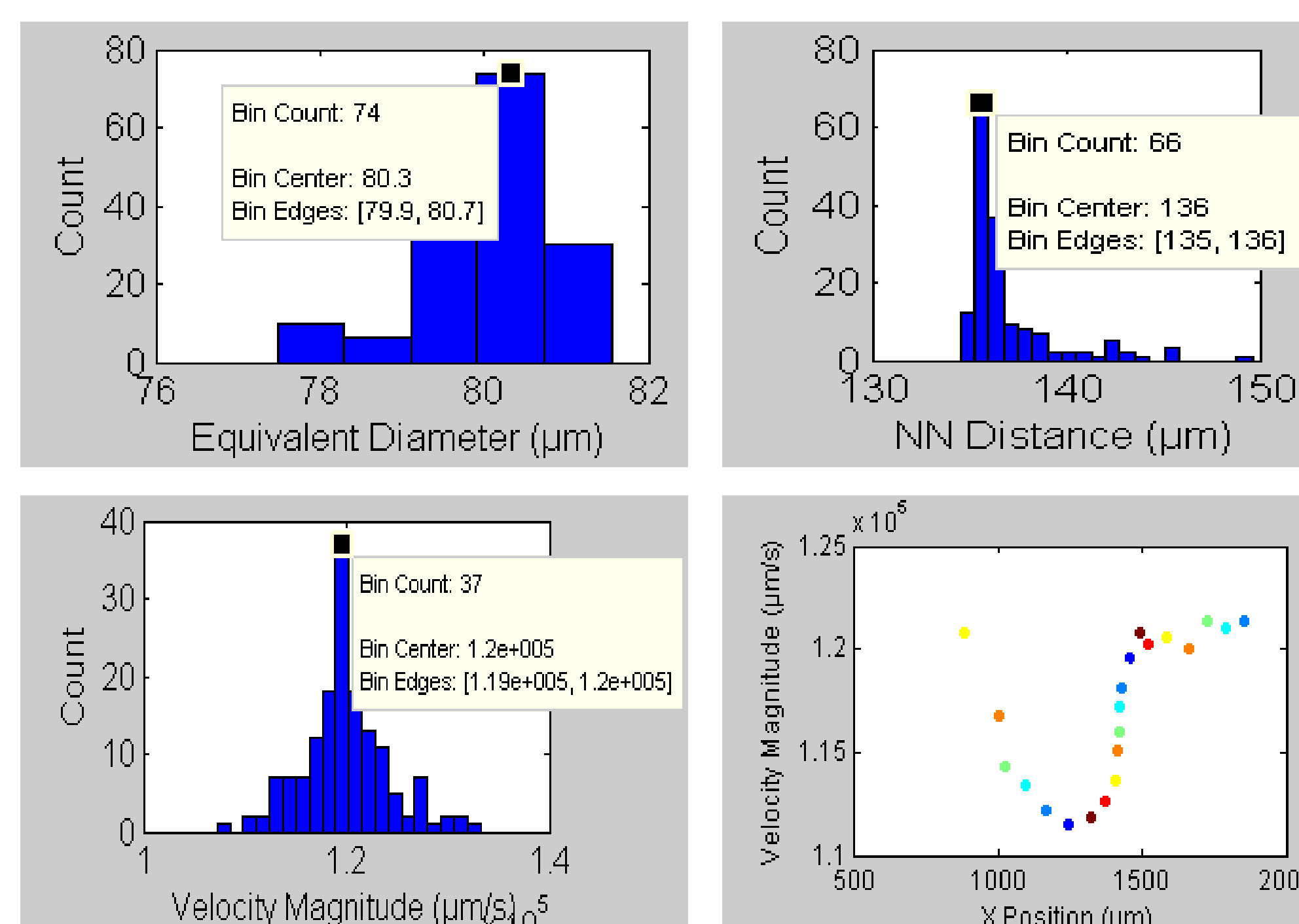
Thanks to a video processing software for time-resolved, label-free tracking of droplet parameters^[4], we determined experimentally the characteristics of the generated flow.



Photron AX100, recording at 6.000 fps, replay at 30 fps

$P_{\text{dodecane}} = 143600 \text{ Pa}$, then $Q_{\text{dodecane}} = 2,07 \text{ mL}\cdot\text{h}^{-1}$
 $P_{\text{water}} = 153000 \text{ Pa}$, then $Q_{\text{water}} = 7,22 \text{ mL}\cdot\text{h}^{-1}$

[4] Basu, A. Lab Chip, 2013, 13, 1892-1901.



Droplet volume comparison between the numerical model based on equations from [2] and [3] and experimental results for the water/dodecane chemical system.

Based on previous works from Van Steijn and Xu, a numerical model was developed to accurately predict the characteristics of droplet production at a T-junction (volumes, frequency, spacing, and specific interfacial area). This numerical model was verified experimentally by generating on-chip segmented flows, and offered ways of improvement in both operating parameters and chip design. Further works will focus on porous hydrophobic membrane (PTFE) phase separation and on implementation of droplet generation for relevant nuclear chemical systems. This work will enable us to characterize each phase during liquid-liquid extraction in order to take into account both matter exchange and hydrodynamics in following mass transfer studies.