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Prediction of the formation of segmented flow in microsystems for radiochemical liquid-liquid extraction

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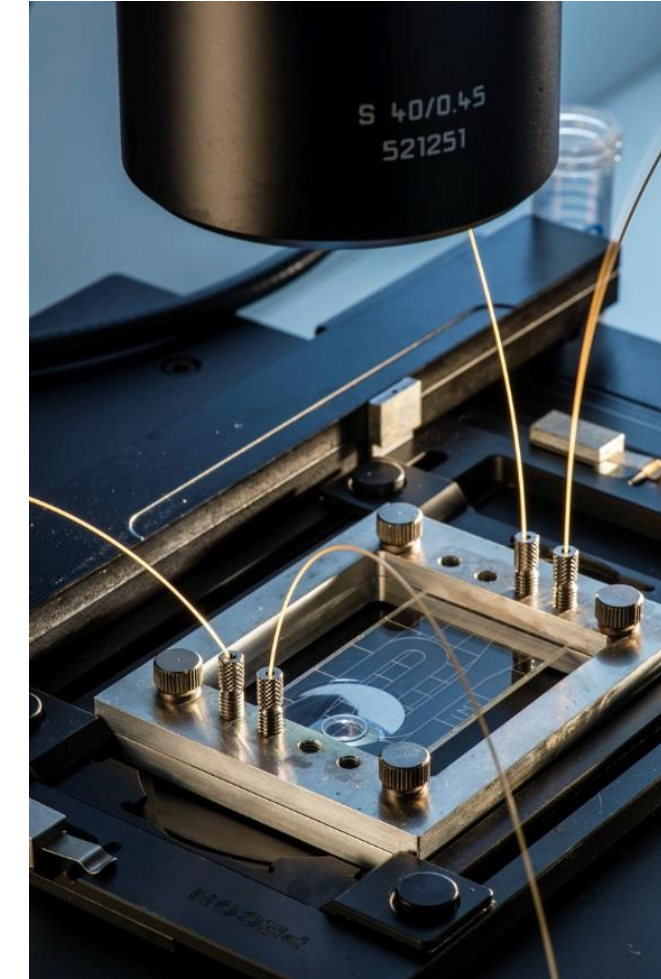
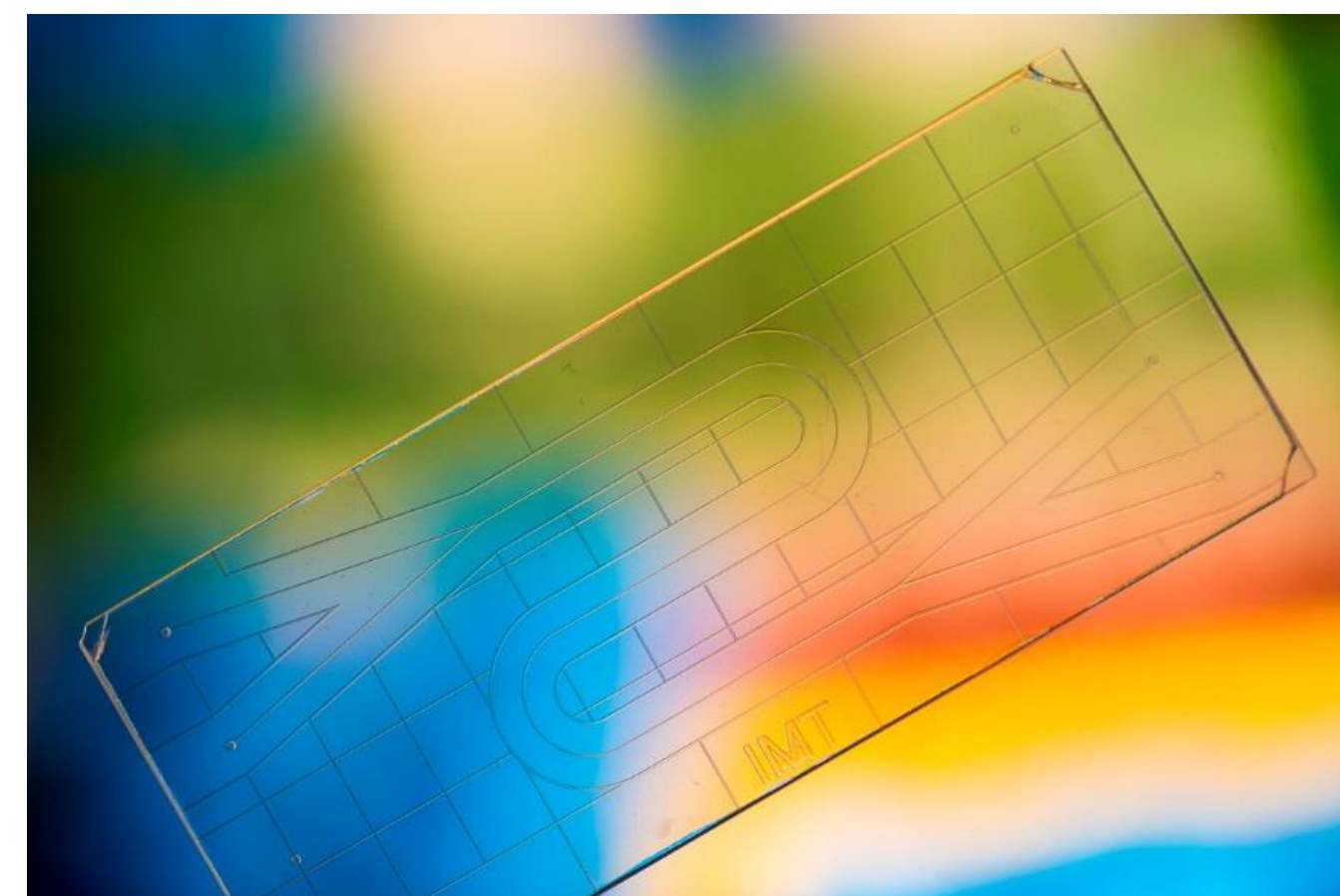
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One of the most important separation techniques in radiochemical procedures is solvent extraction. It involves concentrated acids and corrosive solvents that require microsystems built with robust materials. In the last decade, a growing interest in 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. After a study of parallel flow [1], our goal is to understand and compare the formation of segmented flow for T, flow focusing, and co-flow junctions, and to predict the optimal design in order to enhance solvent extraction.

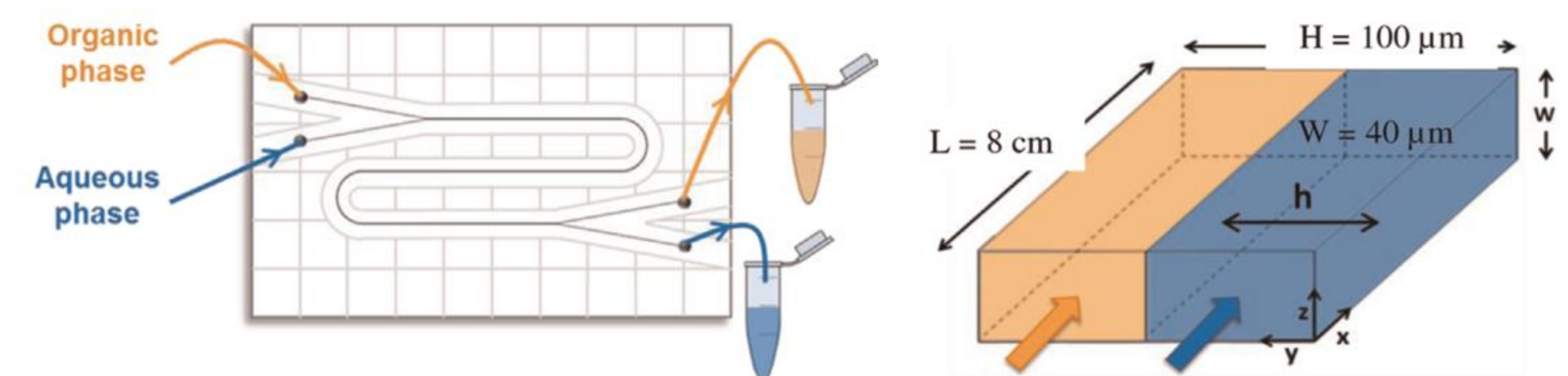
OVERVIEW

Advantages of microsystems

- Volume reduction (from mL to μ L)
- Low operators' exposure to
 - chemicals
 - radiations
- Short analysis time
- High efficiency
- Cost reduction



Parallel flows are mainly used for solvent extraction in microsystems because of a good interface area control and phase separation [1].



Limitation of parallel flow:

- Interface area cannot be optimized
- Liquid-liquid extraction controlled only by diffusion

Not suitable for **low kinetic systems**.

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

DROPLET MICROFLUIDICS

Unlike parallel flows, droplets' mass transfer kinetics are involving both diffusion and convection

1. Enhance the surface accessible for mass transfer to happen by using droplets with a small spacing in between

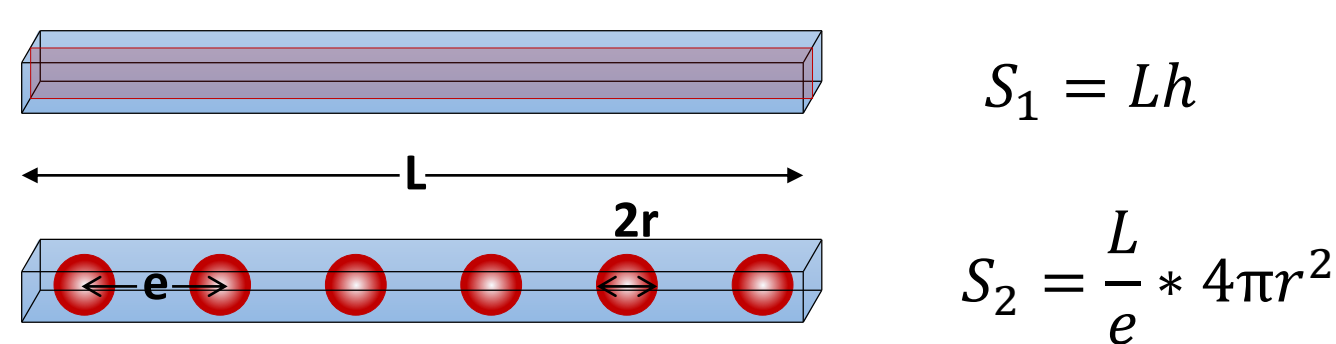


Fig. 1 : Interface area comparison between parallel flows and droplet-based microfluidics

2. Improve mass transfer physically by recirculating species inside droplets

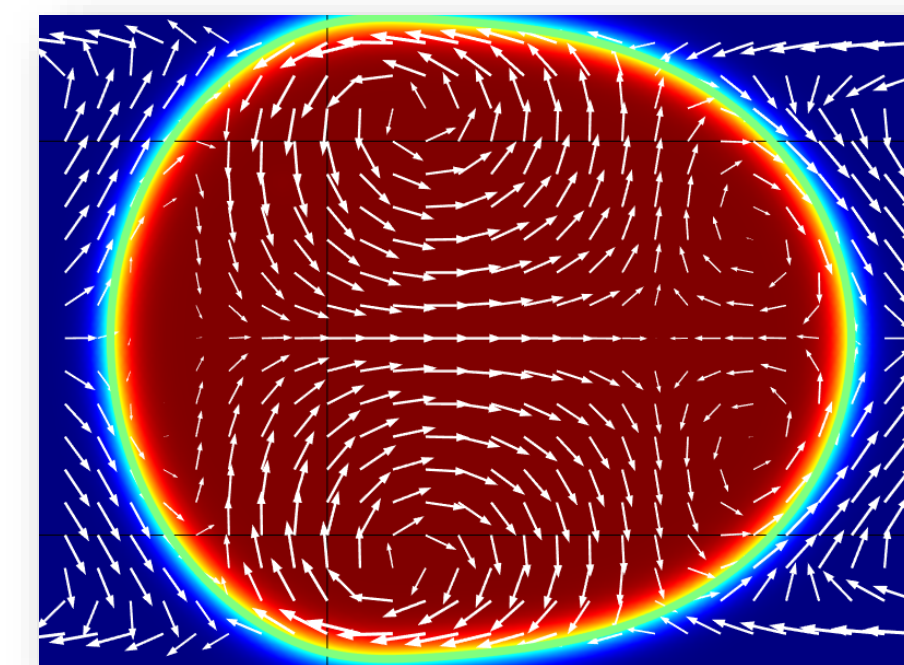
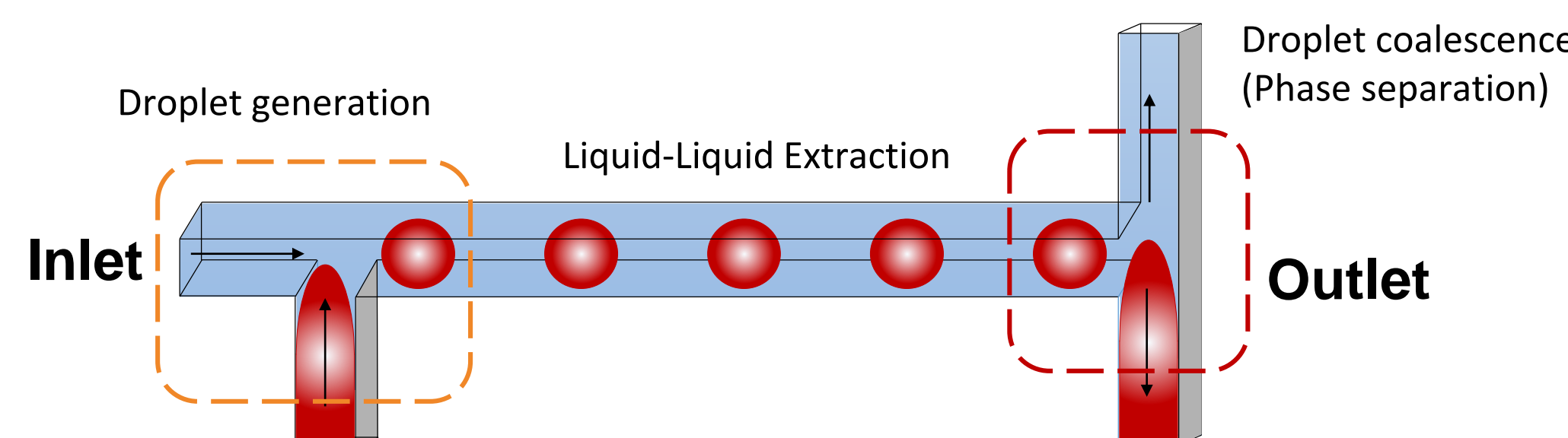


Fig. 2 : 2D CFD Model showing evidence of species recirculation inside 70:30 (v/v) dodecane/TBP droplets, in a continuous phase of $[HNO_3] = 3M$ / $[U(VI)] = 10^{-4} M$, with a pressure gradient of $0.66 Pa \cdot \mu m^{-1}$.

DROPLET LIFETIME

- 1 Inlet : droplets are created by intersecting a continuous and a dispersed phase at a junction



- 2 Outlet : phases are separated by taking advantage of the different affinity of the two phases towards hydrophobic materials

1 DROPLET GENERATION

<p>T-junction</p>	<ul style="list-style-type: none"> <input type="checkbox"/> Very common geometry <input type="checkbox"/> Allows high throughput ($>100Hz$) and monodisperse ($<1\%$) droplet generation [2] 	<ul style="list-style-type: none"> <input type="checkbox"/> Difficult to form tiny droplets (smaller than channel size)
<p>Flow-focusing</p>	<ul style="list-style-type: none"> <input type="checkbox"/> Allows double emulsions (i.e. w/o/w emulsions) <input type="checkbox"/> Access to tiny droplets' volume (i.e. sub-micron) 	<ul style="list-style-type: none"> <input type="checkbox"/> Polydispersity <input type="checkbox"/> Satellite droplets
<p>Co-flow</p>	<ul style="list-style-type: none"> <input type="checkbox"/> Very high throughput droplet generation ($>10kHz$) 	<ul style="list-style-type: none"> <input type="checkbox"/> Insertion of a small capillary in another one <input type="checkbox"/> High dead volume <input type="checkbox"/> Satellite droplets

Parameters affecting flow regime, droplet, droplet size, frequency and spacing

- Viscosities, Flow rates of both continuous and dispersed phases
- Interfacial tension between the two phases
- Geometry and dimensions of the junction

2 DROPLET COALESCENCE

<p>Flow-splitters [3]</p>	<ul style="list-style-type: none"> <input type="checkbox"/> Easy way to separate phases <input type="checkbox"/> Based on wettability differences of the two solvents toward separation materials 	<ul style="list-style-type: none"> <input type="checkbox"/> Commonly used as additional separation device <input type="checkbox"/> Suitable for milliflows
<p>Hydrophobic arrays [4]</p>	<ul style="list-style-type: none"> <input type="checkbox"/> Based on exploitation of capillary forces on selected materials <input type="checkbox"/> Efficient phase separation 	<ul style="list-style-type: none"> <input type="checkbox"/> Expensive microfabrication techniques and controlled environment (white room) required
<p>Surface functionalization [5]</p>	<ul style="list-style-type: none"> <input type="checkbox"/> Easiest and most suitable way to get parallel flow via droplet coalescence <input type="checkbox"/> Robustness due to the covalent nature of the functionalization 	<ul style="list-style-type: none"> <input type="checkbox"/> In-situ preparation

[2] Christopher GF, Anna SL, J. Phys. D: Applied Physics, 2007,40(19), R319-R336.
 [3] Kashid MN et al., Ind. Eng. Chem. Res. 2007, 46, 8420-8430.
 [4] Castell OK et al., Lab Chip, 2009, 9(3), 388-96.
 [5] Hibara A et al., Anal. Chem., 2005, 77(3), 943-947.

MODELLING

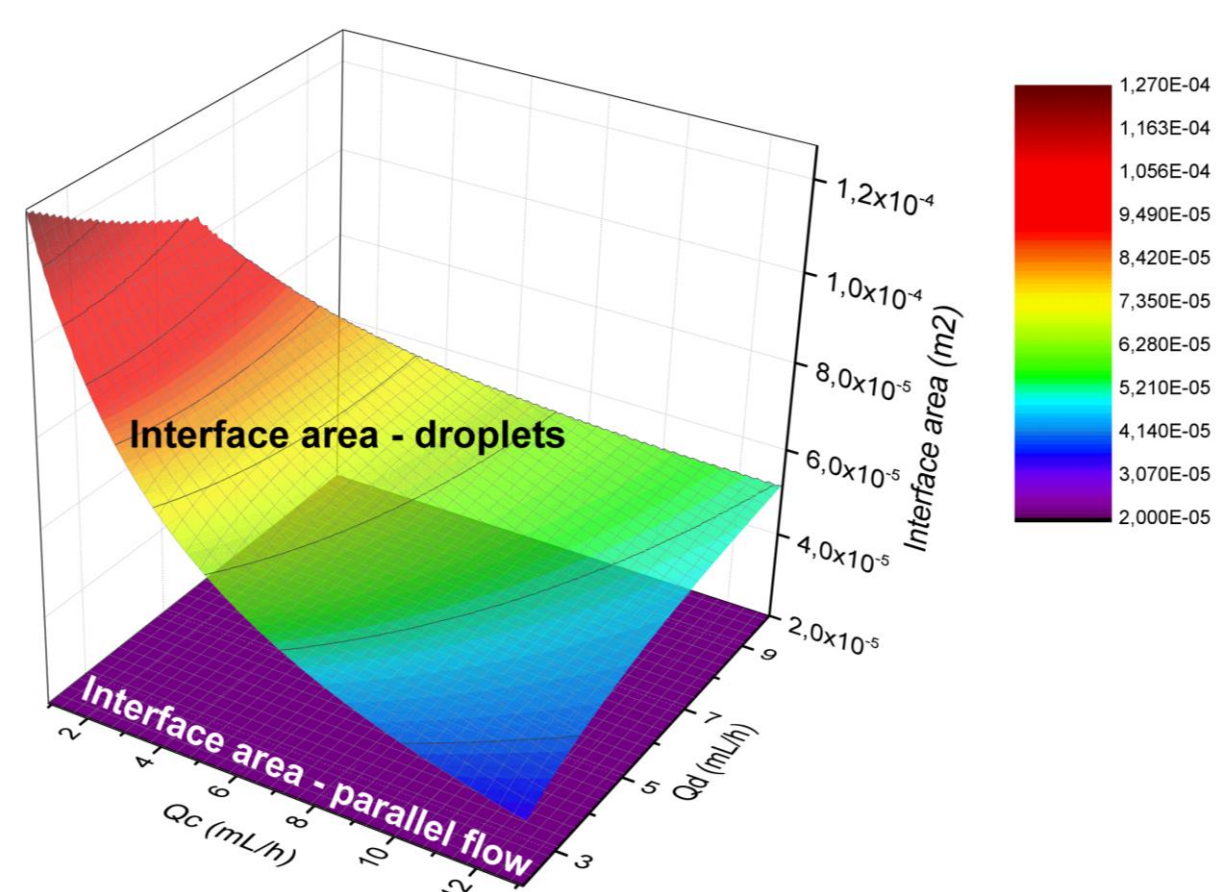


Fig. 3 : Comparison between the interface area generated in a flow-focusing geometry operating in the droplet jetting regime and parallel flow in a similar microsystem [6]

The interface area between the two phases is always greater for droplet-based microfluidics than parallel flows (~5 times higher when optimized).

[6] Utada A S et al., Science, 2005, 308: 537-541.

The size of droplets and their generation rate are shown to be key parameters for liquid-liquid extraction optimization. Microsystems design is therefore investigated in order to determine the operating conditions to control the size and frequency of the droplets, and eventually fasten microsystems prototyping for specific liquid-liquid extractions.

Modelling is an essential asset for rapid prototyping of microsystems in expensive materials resistant to acids and corrosive solvents used in radiochemistry.

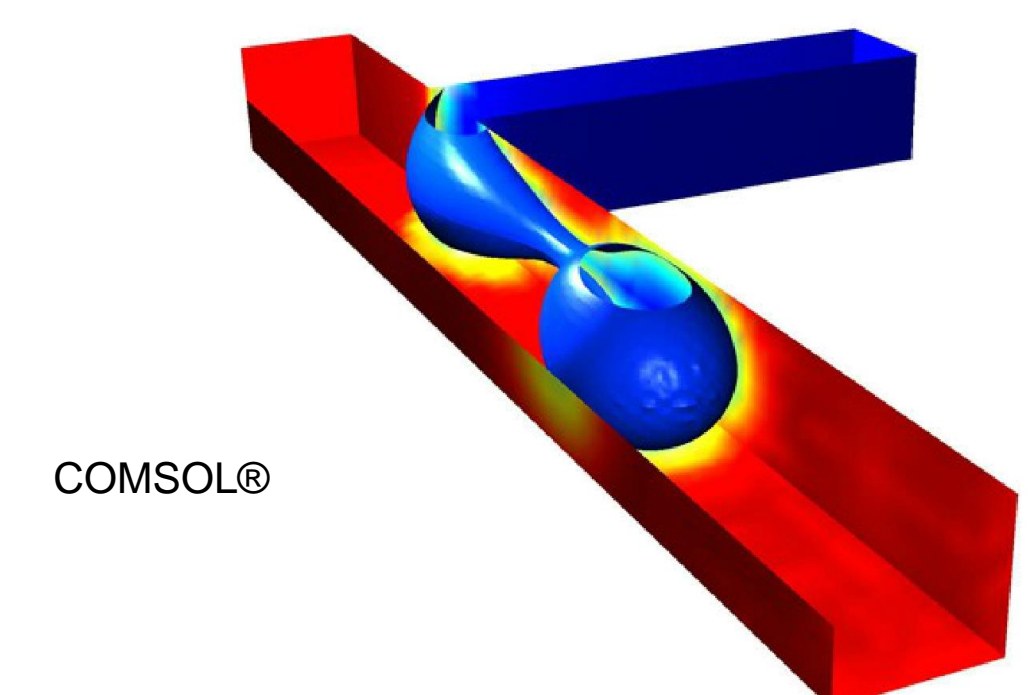


Fig. 4 : CFD simulation of multiphase flow in a T-junction Video accessible online at : https://www.comsol.fr/model/download/11070/droplet_breakup.swf

Using Computational Fluid Dynamics (CFD) models, our future work will be to determine the influence of viscosities, dimensional parameters and operating conditions on flow patterns.

In droplet-based microsystems, the increase in interfacial area and the internal circulation stimulated within the droplets by their passage along the channel are responsible for a large enhancement in the interfacial mass transfer. Both diffusion and convection have an impact on mass transfer, while diffusion was the only means for parallel flows. Therefore, an improvement in liquid-liquid extraction yield might be expected using segmented instead of parallel flows.