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# A coupled experimental / numerical approach for fluids mixing study under supercritical antisolvent process conditions in microreactors

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## 1. Introduction

Supercritical antisolvent techniques have demonstrated promises for processing organic materials at the nanoscale. However, their industrial development is still limited by the poor understanding of the inherent coupled physico-chemical mechanisms (thermodynamics, hydrodynamics, and nucleation-growth). Previous work has demonstrated that it was possible to implement Supercritical AntiSolvent processes in microfluidics devices ( $\mu$ SAS), but without deeper investigations into the physico-chemical phenomena [1]. Indeed, micromixing could have a significant effect over particles size and size distribution since homogeneous concentration distribution and high degree of supersaturation can only be reached by intense micro-mixing obtained through various strategies of mixing geometries. Therefore, we have investigated coflowing fluids at high pressure in a microchip to address the classical limitations and to well control the process conditions. By comparing the experimental observations to the numerical simulation, fluid flow behaviour has been studied for microfluidic mixing and the process condition effects have been captured.

## 2. Results and discussion

In the particular case of miscible fluids conditions ( $\text{CO}_2$  as antisolvent and ethanol as solvent), thermodynamics plays an important role over hydrodynamic behavior. Depending on the considered conditions ( $p$ ,  $T$ ,  $X_{\text{EtOH}}$ ) and the location within the EtOH- $\text{CO}_2$  phase diagram, several different behaviors can be observed. For monophasic mixtures (liquid or supercritical), no obvious interface could be noticed, meaning the mixture velocity plays an essential role to characterize the fluid behavior. By applying a micro Particle Imaging Velocimetry ( $\mu$ PIV) techniques inside microfluidic systems, to a mixture of ethanol and  $\text{CO}_2$  - at SAS conditions – the fluid mixture velocity field is obtained and compared with numerical simulation in order to provide interesting data including the micromixing time. Once the numerical model has been validated by the experimental measurements, it can be used as a valuable tool to study fluid mixing under  $\mu$ SAS conditions. Both laminar and turbulent flow have been examined for different temperatures and  $\text{CO}_2$  ratio in the mixture and their effects are discussed on the segregation intensity, which is a well known criterion of mixture homogeneity. Finally, a characteristic mixing time constant has been defined to evaluate mixing quality and this time constant can be correlated to the energy dissipation rate.

## 3. Conclusions

In order to control a fast mixing of antisolvent and solvent for SAS precipitation, the presented study is focused on the mixing phenomena in microfluidics. An experimental / numerical approach has been proposed: the velocity field of the mixing fluid was measured by  $\mu$ PIV, and the experimental results were used to validate our CFD model. By analysing the simulation results, temperature and  $\text{CO}_2$  ratio effects have been discussed. The mixing time constant is an important term to characterize mixing performance and this study leads to a process optimization to obtain nanoparticles precipitated through SAS process.

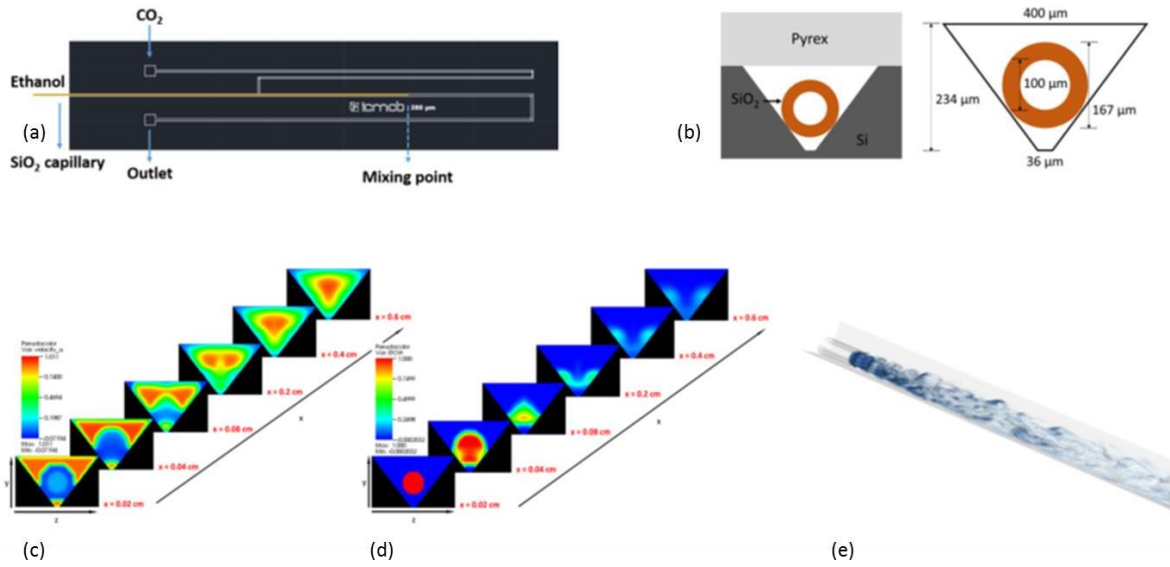


Figure 1. (a) Microfluidic Silicon -Pyrex chip, (b) Cross-section at the mixing point, (c) Velocity component  $u$  evolution in fluid flow direction of a laminar flow, (d) Ethanol field evolution in fluid flow direction of a laminar flow, (e) Turbulent mixing simulation of  $\text{CO}_2$  and ethanol.

## References

- [1]. R. Couto, S. Chambon, C. Aymonier, E. Mignard, B. Pavageau, A. Erriguible and S. Marre, *Chem. Commun.*, **2015**, 51, 1008–1011.