

Slug Flow Capillary Microreactor: Hydrodynamic Study

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Introduction

Microfluidics applications have served as interface between the macro- and nano-world. The physical behavior of fluids (both liquids and gases) at the microscale is a challenging issue for the growing microfluidic applications demands. Microscale systems offer many advantages such as minimal substances consumption, complex chemical waveforms, and significantly reduced analysis or experiment time (for example, an important concept recently introduced was μ TAS, the Micro Total Analysis System, for details see Manz, 1990). The absence of inertial and turbulent effects in microfluidic devices, due to the low ratio between the inertial forces and the viscous forces offers new horizons for physical, chemical and biological applications. Also the short length scales gives high surface-to-volume ratios, small diffusion distances and easy temperature profiling where needed, giving the opportunity to manipulate substances in a better and reliable way. Other significant forces are the surface tension of the fluid and the wetting properties, which takes a very important role at these small scales.

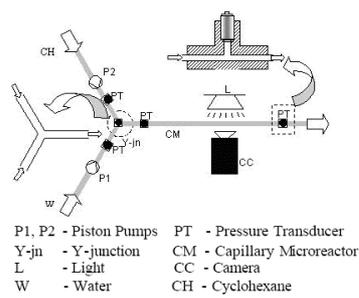
Flow of drops or slugs in small geometries have several medical applications such as: cell-based assays (Pihl, 2005), models for capillary blood vessels for red cells infected with malaria (Shelby, 2003), drug delivery targeted at specific sites in the body for a less invasive chemotherapy, miniature biosamples preparations on fully automated biochips, for DNA sampling and other genomic applications. In addition, it has already been used for different chemical applications in two-phase chemical reactions (Tice, 2004; Hodges, 2004; Harries et. al. 2003), also for kinetic measurement in enzymatic reactions (Song and Ismagilov, 2003), rapid reactions (Burns and Ramshaw, 2001) mass transfer limited reactions, nitration reactions (Burns and Ramshaw, 2002 and Dummann et. al. 2003). Dummann et. al. elucidated and optimized the nitration reaction and achieved significant reduction in byproduct formation using capillary microreactor. Further, easy temperature profiling along the dimensions of the reactor allows carrying hazardous reaction. These microscales reduce the problems associated to the scaling-up for large scale production by simply numbering-up; this means that several microreactors can be used to obtain the necessary products, instead of building complicated and expensive plants which reduces the scale-up risk.

Microreactor technology is relatively a new area and very few mathematical models have been developed to study hydrodynamics of two phase flow (mostly gas-liquid) in small geometries. No model has been developed for liquid-liquid flow in small channel with interface movement and fluid flow movement in its vicinity. In one of our previous works (Kashid et. al. 2005), we have carried out single phase flow simulations to study the internal circulations while in other study (Kashid et. al. 2006a), we carried free surface simulations to understand the mechanism of slug flow generation. In connection with this, there is a need to develop a numerical model which can give detailed information about the two phase liquid-liquid flow in small channels and complex flow in the vicinity of the interface. This task requires powerful modeling techniques; therefore we initiated the hydrodynamic study of drops/slug movement through capillaries and this is what we present in this article.

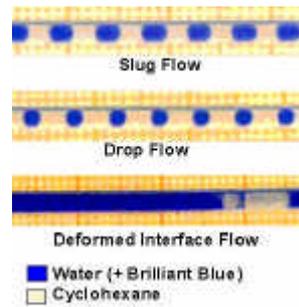
Main results

Experimental results

In Figure 1 a) our experimental setup is presented in which two immiscible liquids (aqueous and organic from two reservoirs) were introduced by continuously operating high-precision piston pumps (throughput range of 1-999 ml/hr) to a symmetric 120° Y-piece mixing element made of Teflon (PTFE). Digital photos were taken with a commercial camera (Olympus E-20P with Macro extension lens WCON-08B) fitted at a length of 0.5 m downstream of the mixing element and a light source (2000 Watt). Though several internal capillary diameters have been tested, in this work we are presenting those made with a 1 mm. In order to distinguish the two phases, the water phase was stained with a blue dye to appear darker than the colorless cyclohexane (see Figure 1 b). The snapshots show that the water phase forms convex shaped slugs, while cyclohexane exhibits a concave geometry as would be expected with the hydrophobic PTFE wall material. The exact form of the slug depends on the inlet flow ratios and the capillary and Y-junction dimensions.



a) Experimental setup scheme

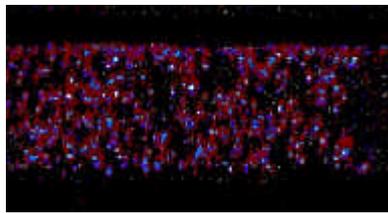


b) Observed flow regimes in the capillary microreactor

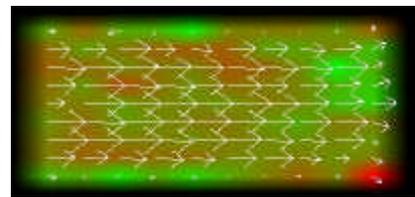
Figure 1. Scheme and photographs

One can easily recognize three well-defined distinct flow regimes: *Slug Flow*, *Drop Flow* and *Deformed Interface* and the detailed explanation of these regimes is given in (Kashid et. al. 2005).

Particle Imaging Velocimetry (PIV) measurements were conducted to gain insight of the internal circulation inside a slug (see Figure 2). And a recirculation pattern was observed and it will be discussed below.



a) Experimental snapshot



b) Velocity distribution of PIV

Figure 2. Internal circulation information from PIV measurements

In the following table, some values of the properties and non-dimensional numbers that characterize this experimental setup are presented:

Table 1. Physical properties and non dimensional numbers.

Property	Cyclohexane	Water phase	Units/Explanation
Viscosity μ	0.001	0.001	kg/m.s
Density ρ	780	1000	kg/m ³
Internal Tube diameter	1.0E-3		m
Surface tension σ	0.05		N/m
Capillary (Ca)	$Ca = \frac{\rho U^2}{\sigma}$	7.06E-5	Viscous over interfacial tension stresses
Reynolds (Re) Slug Diameter	$Re = \frac{\rho U D}{\mu}$	3.53	Inertial effects over the viscous effects
Viscosity ratio (?)	$I = \frac{\mu_{slug}}{\mu_{carrier}}$	1	Slug's viscosity over the carrier's

A complex flow develops in each phase and at the interface between both phases, and this is one of our objectives: *to gain insight of the flow pattern.*

Computational modeling

The 2D hydrodynamic flow pattern of a slug flow in capillary and the evolution of liquid-liquid interfaces were studied by using the in-house developed, open-source, CFD code *FEATFLOW* (see www.featflow.de) with a free surface modeling levelset approach. This code uses an implementation of surface tension effects in interfacial flow combining two techniques: the continuum surface force (CSF) method and a finite element discretization together with the Laplace-Beltrami operator (Hysing, 2006), (Hysing and Turek, 2005), (Turek, S., 1999) and (Turek and Becker, 1998). For the discretization of the domain, the in-house developed software *DeViSoR 2.1* (Design and Visualization Software Resource) was used (Becker and Goeddeke, 2002). A structured two-dimensional coarse

grid was built, see Figure 2 a). The post-processing was performed with *GMV* software (GMV, 1999), and for particle tracing calculations, another in-home module, *gmvpt* (Acker and Turek, 2000), was used.

Essentially, the Level Set method employed solves the Navier-Stokes equation:

$$\mathbf{r}(x) \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) = -\nabla p + \nabla \cdot (\boldsymbol{\mu}(x)(\nabla \mathbf{u} + \nabla \mathbf{u}^T)) + \mathbf{r}(x)g$$

$$\nabla \cdot \mathbf{u} = 0 \tag{1}$$

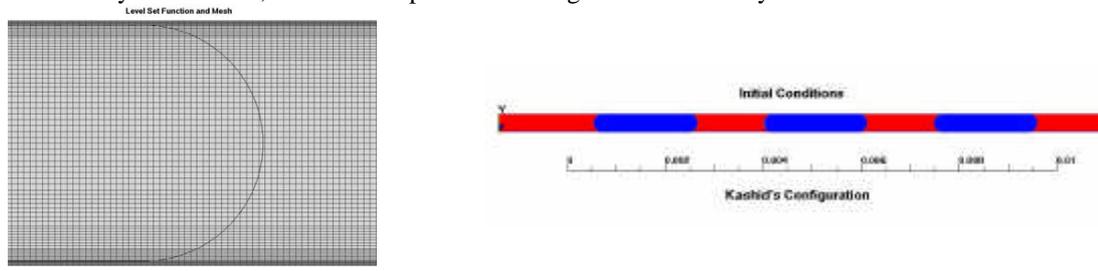
in $\Omega \subset \mathbb{R}^2$, $x \in \Omega$ for varying density ρ and viscosity μ fields and g represents external forces such as gravity.

At the 1D the dimensional interface $\Gamma \subset \Omega$ separating the different fluids, the following boundary condition applies:

$$[u]_{\Gamma} = 0, \quad -[pI + \boldsymbol{\mu}(\nabla u + \nabla u^T)]_{\Gamma} \cdot \hat{n} = sk \hat{n} \tag{2}$$

Where s is the surface tension value; k is the curvature of the interface (Level Set Function) and lastly \hat{n} is the normal vector to the surface interface (for more details see Hysing, 2006). The simulations were carried out on a Sun-Fire-880 computer system with a 900 MHz Sparcv9 processor.

A region of the mesh and the initial configuration of the slugs shape are given in Figure 3. The boundary conditions are: Poiseuille parabolic profile at the inlet and atmospheric pressure at the outlet. Among the main numerical results we have the velocity distributions, which can be plotted as color gradients or velocity vectors.

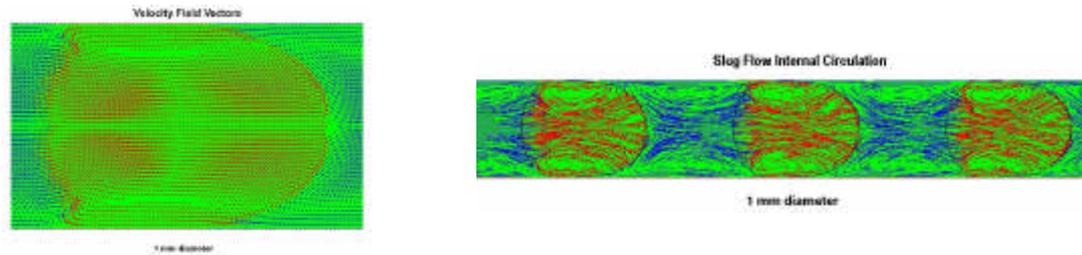


a) Grid and levelset function shape

b) Blue stands for water and red for cyclohexane

Figure 3. Discretization and Initial conditions of both liquids

Detailed information can be extracted of those places where hydrodynamic conditions are the most complicated, e.g. at the interface region, at the nose and back of the slug, etc., see Figure 4.

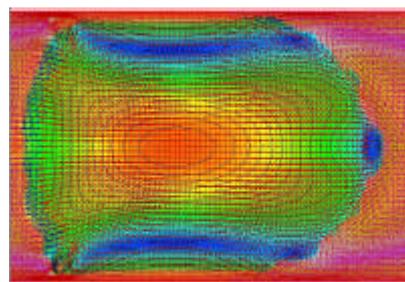
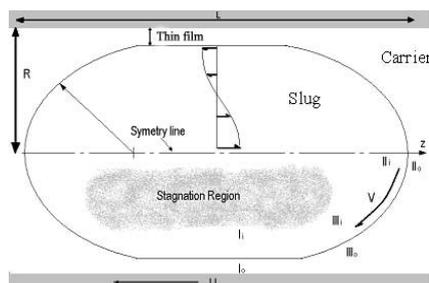


a) Vector plot representing the internal circulation

b) Velocity vector plot of a single slug

Figure 4. Velocity field results at different time steps

As can be seen in Figure 4, there are four stagnation regions that can be identified inside each slug on the upper and downer part and at the front and back ends. Moving in a reference system with the slug's average velocity, the geometry of this configuration provides a symmetry line along the axis of the capillary duct, with two major closed streams of fluid moving like sketched in Figure 5. The centreline of the slug flows in the direction of the slug motion and is formed by the convergent streams of two eddies that separate at the nose (Region II) and go in opposite directions afterwards, reuniting at the tail. This internal convective movement is generated by the shear stress deformation acting on the interface of the slug and the carrier and can be also extrapolated to the internal movement of the carrier (the "i" and "o" subscripts stand for the inside and outside of the slug). In Figure 5 b) the red value stands for the maximum velocity values as for the blue ones mark the stagnation regions (the carrier region is pink shadowed). It was seen that increasing the average carrier velocity, leads to an increase in the internal circulation inside the slugs.



a) Schematic slug regions

b) Contour lines and velocity gradients and vectors

Figure 5. Slugs with the observation point moving with the average slug velocity.

When considering mass transfer and reactions in a microreactor, the transport of the species within the phases will be determined strongly on the hydrodynamic flow pattern. Particle tracing results shows how a possible species can be distributed as time passes by. Initially, a distribution of particles is placed in the back part of the slug, and as the slug moves to the right, the behaviour described above is exactly obtained, see next figures:

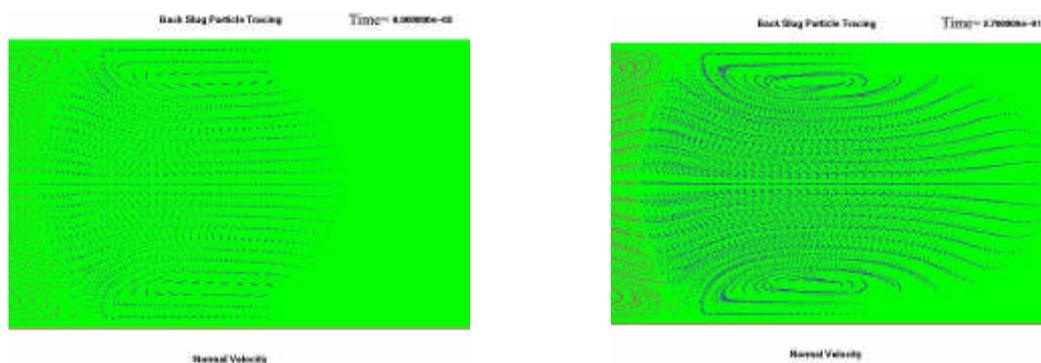


Figure 6. Particle tracing evolution at different times inside a slug.

Comparing the particle tracing numerical solution with the experimental PIV measurements, both results are qualitatively speaking the same and also coincide with the internal circulation expected flow pattern.

Conclusions

The expected experimental physical behavior and numerical results show excellent agreement. The flow pattern occurring inside a biphasic capillary microreactor was successfully studied, experimentally and by numerical modeling. The developed numerical model can be tested under different flow conditions and fluid properties, being a versatile tool for the designing and exploiting of capillaries microreactors. In this way, it will be possible to have a design tool to make sensibility analysis and optimization studies, with the corresponding saving in chemical reactants and laboratory activities which are very expensive. Also many tests can be conducted in parallel in several computer machines, saving also time for finding the required combination of properties, geometries, and many more.

An important issue to be addressed in future works should be the study of the thin film around the aqueous slug, which is a very complex phenomenon. The model tested will be used in future investigations for combining the slug flow hydrodynamics to mass transfer and reaction in such peculiar chemical reactors. Summing all together, the precise control of capillary multiphase flow microreactors, from the hydrodynamics and chemical reaction necessities, will no longer be impossible once this model is validated and tuned.

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