Simulation of Droplet Formation in Micromixers

S. Hardt, F. Schönfeld, F. Weise, Ch. Hofmann and W. Ehrfeld

Institut für Mikrotechnik Mainz GmbH, Carl-Zeiss-Str. 18-20,
D-55129 Mainz, Germany, hardt@imm-mainz.de

ABSTRACT

Droplet formation in micromixers developed by the Institute of Microtechnology Mainz (IMM) is investigated by methods of computational fluid dynamics (CFD). The CFD results are compared to experimental data obtained with transparent micromixers made from photosensitive glass. Preceding the 3D simulations, the decay of liquid cylinders is studied as a test case. Subsequently, the flow patterns of a binary system of immiscible liquids in a interdigital micromixer are investigated. It is found that in case of equal volume flows the simulation results on the decay wavelength and on droplet and lamellae diameters agree very well with experimental data. However, the agreement between simulations and experiments is less convincing in the case of a 1:4 volume flow ratio. Further and more detailed studies for a broader range of operation and material parameters are ongoing.

Keywords: Micromixers, Droplet formation, Free surface flows

1 INTRODUCTION

Emulsions and dispersions are of considerable importance as intermediate and final products of the chemical industry. The application spectrum of disperse phases extends from magnetic thin films to pharmaceuticals. The formation of an emulsion or a gas-liquid dispersion requires an energy input to increase the interfacial area, which is usually provided by mechanical actuators such as stirrers or centrifuges. Micromixers offer an alternative method for emulsion and dispersion generation in a continuous flow process [1]. Typically, in micromixers a fluid stream is split into several substreams. Subsequently, the substreams corresponding to the disperse phase decay into droplets or bubbles, thus forming an emulsion or dispersion, respectively.

Micromixers offer several advantages over conventional mixing technology. Under certain conditions a very narrow droplet size distribution is found [2]. Furthermore, emulsion formation in micromixers requires less energy than the corresponding process in a stirred tank reactor [3].

One of the most frequently used micromixers developed at the Institute of Microtechnology Mainz (IMM) is the so-called interdigital mixer. In such a type of mixer fluid lamellae are created by an interdigital arrangement of inlet channels with alternating feeds, as shown in Fig. 1. The width of fluid lamellae created with the interdigital feed structures typically lies between 25 and 40 μm.

In order to perform flow distribution measurements, a couple of different interdigital mixers were fabricated by the Mikroglas company, Mainz, from photosensitive glass by means of a UV-lithography process. In contrast to Fig. 1, a planar design was chosen, where channels for inlet and mixing streams are arranged in one layer. The transparent cover and bottom plates allow to examine the flow distribution in the mixing zone by transmitted light microscopy.

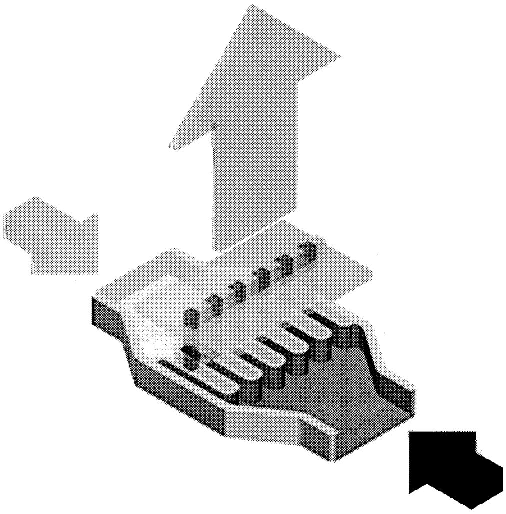

Figure 1: Schematic design and flow scheme of an interdigital mixer.

2 DECAY OF LIQUID CYLINDERS

Using the glass micromixers, experiments were performed with feeds of silicon oil and water forming an emulsion. The modeling and simulation of emulsions in microfluidic devices is a computationally challenging task. As opposed to macroscopic vessels, droplet or bubble diameters are often comparable to characteristic system dimensions. For this reason, it is necessary to spatially resolve the phase boundary and transient simulations describing the time evolution of the interface are required.

Most suitable for modeling complex 3D free surface flows with topology changes are volume-tracking techniques [4,5]. The basic idea of these approaches is to define a function *f* describing the local volume fraction of a specific fluid. The value of *f* is 1 within the corresponding fluid and 0 outside. The volume fraction obeys an advection equation given as

$$\frac{\partial f}{\partial t}+u_i\frac{\partial}{\partial x_i}f=0 \tag{1}$$

When solving Eq. (1) based on a spatial discretization method such as FDM or FVM, discretization errors are introduced effecting a diffusive smearing of the volume fraction field. Thus, without further precautions an initially sharp interface becomes blurred, making it finally impossible to identify the spatial regions belonging to different phases. This problem can be solved by numerical techniques allowing to reconstruct the exact position of the interface within computational cells [4]. These so-called volume-of-fluid (VOF) methods have proven to be powerful but computationally intensive. In order to reduce the computational effort for the 3D problems to be solved a somewhat simplified approach is followed in the present work. The approach relies on a correction algorithm reversing interface smearing by numerical diffusion. The algorithm determines the fluid mass which has penetrated the interface in a certain time step, removes the mass from the wrong side of the interface and adds it to cells where fluid has been depleted. This so-called "surface-sharpening" algorithm, implemented in the finite volume solver CFX4 of AEA Technology, ensures *global* mass conservation. However, when the fluids form several disconnected regions, *local* quantities as the mass of a specific droplet may not be conserved.

The volume tracking method described above is supplemented by a continuum method for modeling surface tension [6]. Due to the large surface-to-volume ratio, surface tension plays an important role in microfluidic systems and cannot be neglected.

Frequently, droplet or bubble formation occurs via a hydrodynamic instability. For reasons that will become clear in the following section, some of the liquid lamellae created in the interdigital mixer get rearranged in a cylindrical shape. Before studying the 3D flow problem of the micromixer it is thus advisable to first investigate the axisymmetric problem of a decaying liquid cylinder. The decay of a cylindrical liquid jet has been studied intensely by both experimental and theoretical methods [7]. Therefore, besides its relevance for emulsion formation, this problem serves as a test case for the numerical methods to be used.

Since the groundbreaking work of Rayleigh [8] it is known that an infinitely long liquid cylinder is unstable under small perturbations of its surface shape. The so-called Rayleigh-Plateau instability sets in at infinitesimal sinusoidal modulations of the surface radius R with a wavelength greater than $2\pi R$. Linear stability theory for inviscid, irrotational flow predicts a maximum growth rate for perturbations with a wavelength of $9.01R$.

In order to investigate the Rayleigh-Plateau instability and the further time evolution of the system when nonlinear effects become important, an axisymmetric 2D model was set up. As an initial state, sinusoidal modulations were superposed to a liquid cylinder of 100 μm radial and 1.8 mm axial extension. Symmetry boundary conditions were used at $z = 0$ and $z = 1.8$ mm. The cylinder consists of water and is surrounded by silicon oil, which has a viscosity more than 9 times larger than that of water. In Rayleigh's original work the influence of the outer fluid has been neglected. For this reason, the case of a water cylinder surrounded by air was studied additionally.

In the simulations, performed with the finite volume solver CFX4, both two-fluid systems showed a qualitatively similar behavior. Fig. 2 shows the time evolution of the water/silicon oil system for an initial perturbation with a wavelength of $4.5R$. As expected from linear stability theory, the subcritical perturbation is damped and a cylindrical shape is recovered after $t \approx 0.5$ ms. However, before the kinetic energy is dissipated into heat, a mode with $\lambda \approx 9R$ is excited (third frame). Eventually, the cylinder decays into droplets after thin fluid rods have been formed in the pinching regions.

As a common feature of the systems with both a high and a low viscosity outer fluid, the decay into droplets occurs via coupling to excitations of longer wavelength for perturbations below $\lambda \approx 2\pi R$. Even when the initial perturbations lie in the range of linearly unstable modes, significant admixtures of modes with $\lambda \approx 9R$ are found in the final configuration of the system. Thus, for a broad range of initial conditions, the Rayleigh mode governs the dynamics.

Despite artifacts related to small satellite droplets, no major violations of local mass conservation were visible on the time scales considered here. It can thus be concluded that the numerical method used seems to be an appropriate tool for describing the gross structure of droplet formation in binary systems of immiscible liquids.

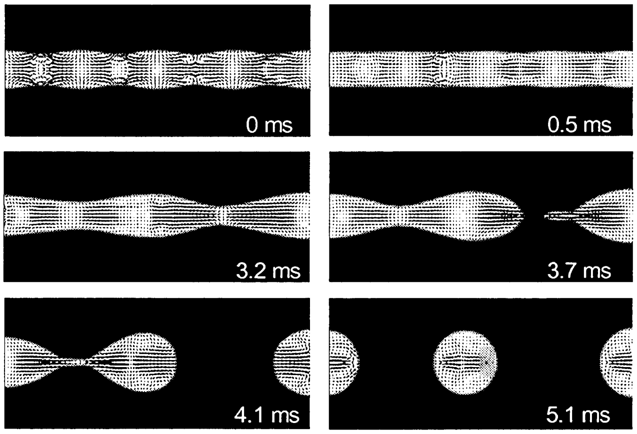

Figure 2: Time evolution of an initially perturbed water cylinder surrounded by silicon oil.

3 DROPLET FORMATION IN MICROMIXERS

Compared to the decay of a liquid cylinder at rest, the dynamics of emulsion formation in a micromixer is more complex. Due to the nonzero velocity of the liquid lamellae created by the interdigital channel structures, shear forces are induced. Furthermore, the lamellae are in contact with the channel walls when entering the mixing zone. For this reason, wetting forces are expected to play a role for the dynamics. Fig. 3 shows photographs of the emulsification process in a transparent interdigital micromixer. The liquids, silicon oil and water, to which a blue dye was added, enter the mixer through 30 inlet channels from the left. At the interface to the mixing zone the inlet channels have a cross-sectional area of 60 μm × 150 μm, separated by 50 μm walls. The cross section of the mixing zone, only a part of which is shown in the figure, is 3250 μm × 150 μm.

The photographs of the mixing zone were taken with a CCD camera. In the top part of Fig. 3 a volume flow ratio of water and silicon oil of 1:1 was chosen, in the bottom part the ratio is 1:4. The total liquid volume flow is 800 ml/h in both cases. Even with the limited spatial resolution of the CCD camera water lamellae and droplets become visible. Whereas in the case of equal volume flows the droplets are formed by lamellae decay, for a flow ratio of 1:4 droplet formation occurs very close to the inlets. Furthermore, it is apparent that in the case of the 1:4 flow ratio the droplets are smaller and the distance between droplets is larger than in the case of the 1:1 flow ratio.

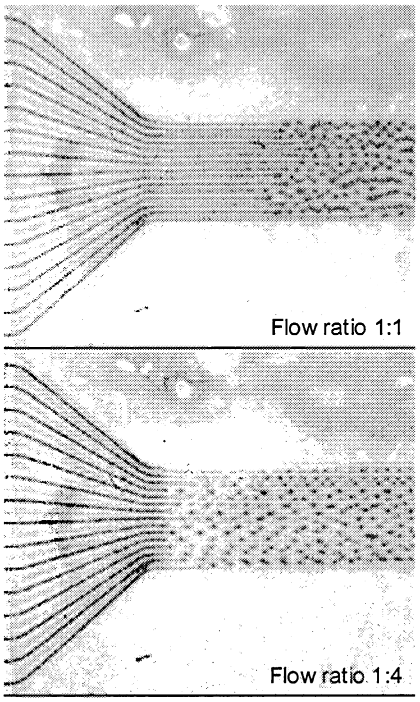

Figure 3: Optical images of droplet formation in micromixers for different flow ratios.

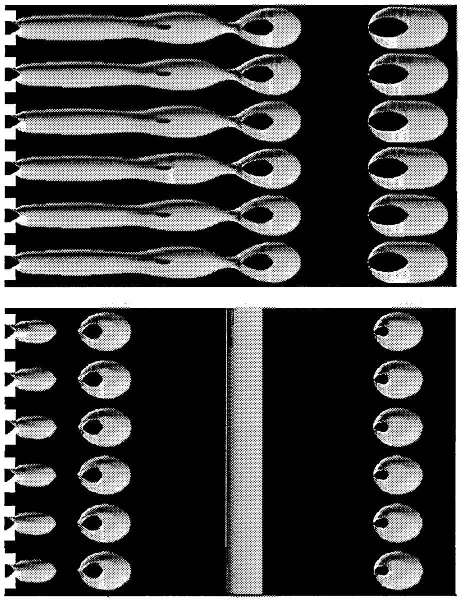

Figure 4: Snapshots of the phase boundary for a flow ratio of 1:1 (above) and 1:4 (below) as obtained from the simulations.

In order to describe emulsion formation in the micromixer, a 3D computational model was set up. Only the region around one single fluid lamella was considered. The remaining parts of the mixer were accounted for by periodic boundary conditions. Furthermore, due to CPU time constraints, in flow direction only a slice of the mixing zone of up to 4 mm length was considered. Since wetting forces are expected to be of major importance, the contact angle of the system silicon oil/water needed to be determined. With the measuring equipment available at IMM no reliable measurement of the contact angle was possible. As it was found that silicon oil has far better wetting properties than water for the glass material chosen, an angle of 30° was used in the simulations. All simulations were performed with the flow solver CFX4 using the method described in the previous section.

Fig. 4 shows snapshots of the water/silicon oil interface for two different flow ratios and a total volume flow of 600 ml/h. For better visibility of the 3D structures, the water lamellae are shaded in light gray, in contrast to Fig. 3. In the case of the 1:1 flow ratio water lamellae are formed, which, by action of wetting forces, detach from the channel walls and assume a circular cross section. The lamellae decay into droplets which again touch the upper and lower channel wall. In the figure, the contact areas at the channel walls are visible as elliptical holes in the liquid interface. In agreement with experimental data, droplet formation occurs close to the inlets in case of the 1:4 flow ratio. Frequently droplet coalescence was observed in the simulations. In middle part of Fig. 4, neighboring droplets have merged and form a liquid column. However, details of the coalescence mechanism are not likely to be realistically described, as, due to the periodic boundary conditions, the time evolution of different lamellae is assumed to be identical. In practice, neighboring lamellae do not decay in a completely identical manner, and coalescence is avoided when a droplet is formed in between two neighboring droplets.

Flow ratio	Lamella diameter [µm]		Decay wavelength [µm]		Droplet diameter [µm]	
	Exp.	Simul.	Exp.	Simul.	Exp.	Simul.
1:1	100	100	470	513	236	236
1:4	-		386	641	158	203

Table 1: Comparison of experimental and simulation results

Experimental and theoretical investigations on droplet formation in micromixers are ongoing and a final analysis is not yet available. Table 1 shows a first comparison of experiments and simulations, using the yet very restricted results database. The evaluated quantities, the average diameter of the water lamellae, the decay wavelength (i.e. the distance between subsequent droplets) and the droplet diameter are, with regard to the dataset available, independent of the total volume flow. The flow ratio, however, seems to have a significant influence on the droplet formation mechanism, as already apparent from Fig. 4. Whereas experimental and simulation results agree very well for the 1:1 flow ratio, major deviations are found in case of the 1:4 flow ratio. A possible explanation for this fact can be found in Fig. 5. In Fig. 5 the length of water lamellae (i. e. the length after which droplets pinch off) is plotted as a function of the single phase volume flow in the case of equal volume flows. The simulation seems to match with experimental results at higher volume flows, but seems unable to correctly describe the transition from droplet formation at the inlets to formation and decay of lamellae. The mismatch in Table 1 could be due to a difference in the droplet formation mechanism: the simulation predicts lamella decay, but in reality droplet formation occurs at the inlet. The transition between different formation mechanisms is expected to sensitively depend on the wetting behavior of the two-liquid system. Thus, more reliable measurements of the contact angle are needed.

When relating the decay wavelength λ_c to the lamella radius R, one obtains $\lambda_c = 9.40R$ and $\lambda_c = 10.26R$ from the experiments and simulations, respectively. As these wavelengths are quite close to the Rayleigh wavelength of $9.01R$, it is natural to assume that the Rayleigh-Plateau instability plays a dominant role for the decay of liquid lamellae in micromixers.

4 SUMMARY AND OUTLOOK

Binary systems of immiscible liquids in micromixers were studied both experimentally and by simulation methods. The Rayleigh-Plateau instability of a liquid cylinder was chosen as a test case for the computational approach of volume tracking.

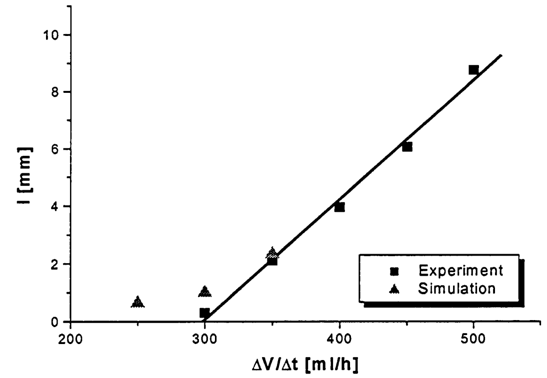

Figure 5: Length of the water lamellae as a function of volume flow.

The simulation results obtained comply with results of a linear stability analysis. Subsequently, simulations of droplet formation in micromixers based on a 3D model were performed. In the case of equal volume flows of silicon oil and water a good agreement between simulations and experimental results is obtained. However, in the case of a flow ratio of 1:4 a discrepancy between simulations and experiments is visible. As suggested by an evaluation of the lamella decay wavelength, the Rayleigh-Plateau instability seems to play a dominant role for droplet formation. The analysis will be continued with more detailed studies for a broader range of material and operation parameters.

Acknowledgements

We would like to thank A. Freitag and T. Dietrich from Mikroglas AG Mainz for their help regarding the design of the glass micromixers and for realiziation of these devices. Furthermore, we gratefully acknowledge the support of M. Eisenlauer from AEA Technology, Germany. This work was supported by a grant from Volkswagen Stiftung.

REFERENCES

[1] H. Löwe, W. Ehrfeld and V. Hessel, Proc. of IMRET4: 4th Int. Conf. on Microreaction Technology, AIChE, 31, 2000

[2] V. Haverkamp, W. Ehrfeld, K. Gebauer et al., Fresenius Journal of Analytical Chemistry 364, 617, 1999

[3] T. Bayer, H. Heinichen and T. Natelberg, Proc. of IMRET4: 4th Int. Conf. on Microreaction Technology, AIChE, 167, 2000

[4] C.W. Hirt and B.D. Nichols, Journal of Computational Physics 39, 201, 1981

[5] W.J. Rider and D.B. Kothe, Journal of Computational Physics 141, 112, 1998

[6] J.U. Brackbill, D.B. Kothe and C. Zemach, Journal of Computational Physics 100, 335, 1992

[7] J. Eggers, Review of Modern Physics 69, 865, 1997

[8] J.W.S. Rayleigh, "The Theory of Sound, Vol II, 2nd ed.", Dover, New York, 1945