Mixing process of immiscible fluids in microchannels

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A R T I C L E   I N F O

Article history:
Available online xxxx

Keywords:
Microchannel flows
Immiscible fluids
Interface
Surface tension
Vortical structures
VOF method

A B S T R A C T

The paper is concerned with experimental investigations and numerical simulations of immiscible flows in microchannels in the presence of interfacial surface tension. The aim of the present study is to model the unsteady dynamics of the vortical structures and interface shape in the Y-branching geometries with square cross-sections. The tested fluids are Newtonian and weakly elastic polyelectrolyte solutions in water (modelled as shear thinning Carreau–Yasuda fluids). The experiments used specially designed configurations based on optical and confocal microscopic devices; PIV measurements of the velocity distributions and direct visualizations of the interface are obtained. The simulations performed with the VOF solver implemented in the FLUENT code are validated by experiments for small and medium Reynolds numbers (0.1 < Re < 200). The numerical solutions offer a detailed description of the vortices formation in the vicinity of the interface, phenomena which directly influence the mixing and diffusion processes within the micro-geometry here investigated.

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1. Introduction

The mixing of fluids in microchannels is an essential hydrodynamic process for many of the lab-on-a-chip applications (Stone et al., 2004; Kim et al., 2008, 2009). Numerous studies have recently been directed at analysing non-homogeneous fluid flows in micro-bifurcations at low Reynolds numbers (Chang and Yang, 2007; Kim et al., 2009; Sato et al., 2007; Tollefberg et al., 2009), with applications to biofluid mechanics (Kersaudy-Kerhoas et al., 2010; Katsumoto et al., 2010), dispersions in microchannels (Adjari et al., 2006) and diffusion–extraction processes (Mata et al., 2008; Pan et al., 2007). In almost all the papers the experimental investigations (Santiago et al., 1998; Huh et al., 2009; Liu and Peng, 2009) are corroborated with numerical simulations (Balan et al., 2007; Fujisawa et al., 2006; Ramirez and Conlisk, 2006) in order to have a better understanding, representation and characterization of the mixing/diffusion processes within microchannels (Adeosun and Lawal, 2009; Chung et al., 2009; Silva et al., 2008; Yamaguchi et al., 2004).

The modelling of mixing flow processes (liquid–liquid or gas–liquid contact phases) is normally associated with diffusion processes (Adeosun and Lawal, 2005; Liu et al., 2004), transport of air bubbles in liquids (Fu and Pan, 2009; Gupta et al., 2009), capillary flows (Zhu and Petkovic-Duran, 2010), wall channel influences (Huh et al., 2009; Yamasaki et al., 2009), flow control (Wang et al., 2007) and Taylor flows (Guo and Chen, 2009; Qian and Lawal, 2006). Despite the large numbers of published papers dedicated to mixing fluids in micro-geometries, only a few studies have focused on the dynamics of the interface between immiscible liquids in microchannels, most of the investigations being concerned with liquid–liquid displacement phenomena and interface instability (Soares and Thompson, 2009; You and Zheng, 2009; Zhao et al., 2006).

The goal of the present study is to obtain a more detailed representation of the vortical structures generated by a moving interface between two immiscible liquids in a Y-micro-configuration, in the presence of interfacial surface tension.

The unsteady vortex pattern in micro-branching 3D-flows is not easily observable by experiments, especially if the average velocity is relatively large (i.e. V > 10 mm/s). In such cases, CFD techniques have to be used to obtain a better description of the local flow kinematics, see Silva et al. (2008). The evolution and distortion of the interface shape near the bifurcation are numerically investigated in this paper using the unsteady volume of fluid (VOF) solver available within the FLUENT code. We also explore the influence of the shear thinning character on the flow structure. The numerical results correlate the pattern of the vortices with material parameters and provide evidence of the velocity fluctuations produced by the travelling of the interface.

The topic is of major interest if a diffusion process is associated with the basic hydrodynamics. In the vicinity of a separation surface, the local kinematics within each phase influence directly the rate of diffusion and mass transfer between the fluids in contact, see also Münchow et al. (2008). In microchannels with an aspect ratio close to one, the liquid–liquid displacement produces...
Nomenclature

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<thead>
<tr>
<th>Symbol</th>
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<tr>
<td>$d$</td>
<td>characteristic channel dimension, m</td>
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<tr>
<td>$n$</td>
<td>shear thinning index</td>
</tr>
<tr>
<td>$Re$</td>
<td>Reynolds number</td>
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<tr>
<td>$V$</td>
<td>average velocity, m/s</td>
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<tr>
<td>$v_z$</td>
<td>velocity component on z-direction, m/s</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>shear rate, 1/s</td>
</tr>
<tr>
<td>$\eta$</td>
<td>shear viscosity, Pa s</td>
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<tr>
<td>$\eta_0$</td>
<td>zero shear viscosity, Pa s</td>
</tr>
<tr>
<td>$\eta_{\infty}$</td>
<td>infinite shear viscosity, Pa s</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Carreau time constant, s</td>
</tr>
<tr>
<td>$\rho$</td>
<td>mass density, kg/m$^3$</td>
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<tr>
<td>$\sigma$</td>
<td>interfacial surface tension, Pa m</td>
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Greek symbols

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Fig. 1. Experiment and numerical simulation of the stationary single-phase flow in Y1-channel ($V = 0.225$ m/s, sample: water, $Re = 160$); (a) flow branching geometry, (b) direct pathlines visualization, and (c) 3D numerical simulation – pathlines in the mid-plane (the closed branch is marked with the cross symbol).

Fig. 2. Evolution of the interface between water and air in vicinity of the junction of Y1-channel: experiment and numerical simulations. The entrance velocity of water is $V = 0.1$ m/s (the closed branch is marked with the cross symbol). Numerical 3D unsteady VOF simulations are performed in the presence of surface tension (0.072 Pa m, wall contact angle of water is 90°).

Please cite this article in press as: Balan, C.M., et al. Mixing process of immiscible fluids in microchannels. Int. J. Heat Fluid Flow (2010), doi:10.1016/j.ijheatfluidflow.2010.06.008
<table>
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<th>Experiment</th>
<th>Numerical simulations</th>
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<td>![Experiment Image]</td>
<td>![Numerical Simulations Image]</td>
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**Fig. 3.** Experiment and simulations of the interface evolution in the Y2-channel between two identical liquids (low concentration of proteins in water), one marked with fluorescein (the entrance average velocities are equal on each branch, i.e. \( \dot{V}_1 = \dot{V}_2 = 0.0012 \text{ m/s} \)). The corresponding 2D unsteady VOF simulations are performed without interfacial tension.

**Fig. 4.** Micro-PIV experimental set-up for measuring the velocity distribution in the Y1-channel.
unsteady 3D-vortical structures in the neighbourhood of an interface, phenomena which induces a local mixing similar to pulsatile flows (Hitt and McGarr, 2004).

2. Validation of numerical simulations

Prior to any qualitative and quantitative analysis of the flow under investigation, the experimental validation of the numerical code is essential. In this paper the computations are performed with the FLUENT 6.3 code using the unsteady laminar Navier–Stokes solver (2D and 3D configurations) and the VOF procedure for the determination of the interface between immiscible fluids (similar computations applied to model the Taylor flow in branching microchannels have been presented by Guo and Chen (2009) and Qian and Lawal (2006)). The investigated flows are in the range of small and medium Reynolds numbers, $0.01 < \text{Re} < 200$ with $\text{Re} = \rho \cdot \text{V} \cdot d / \eta$. Two types of Y-microchannels geometries are used in the present work. The first test geometry, Y1-channel, has a square section of $0.7 \times 0.7 \text{ mm}$; it is symmetric with an angle of $75^\circ$ between the branches, one branch being kept closed during the tests (Fig. 1). Direct visualization and numerical computations

![Image](image_url)

**Fig. 5.** Micro-PIV experimental measurements and numerical simulations of the Newtonian steady flow field in the Y1-channel: (a) numerical flow spectrum at $\text{Re} = 10$, (b) velocity distributions along the main flow direction ($\text{Re} = 10$, PIV measurements), and (c) corresponding velocity distribution upstream the junction, as function of Reynolds number (the used sample is water).

![Image](image_url)

**Fig. 6.** Complex viscosity function of PS sample (polyacrylamide solution in water, molecular mass $18 \text{ M}$, concentration $1000 \text{ ppm}$); the experimental data are modelled with relation (1): $\eta_\infty = 2.7 \text{ Pa s}$, $\eta_0 = 3 \text{ mPa s}$, $\lambda = 7 \text{ s}$, $n = 0.2$, $\alpha = 1$. 

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of the interface evolution between two immiscible fluids are shown in Fig. 2.

The second investigated geometry is the Y2-channel with a cross section of 0.3 x 0.37 mm. In Fig. 3 comparison between experiments and numerical simulations of the interface between two identical liquids is shown.

The results presented in Figs. 1–3 give the confidence in the numerical solutions, both in steady and unsteady mode and a fair qualitative modelling of the interface shape evolution between immiscible fluids.

The measured distributions of the velocity field in the micro-channel are obtained using the micro-PIV DANTEC set-up presented in Fig. 4. The fluid sample is transported at a constant flow rate in the transparent Y1-channel test geometry, the sample being enriched with reflecting particles of 1 micron diameter. The corresponding velocity field is measured on the geometrie mid-plane using the specialized DANTEC software and the corresponding pattern of the vortical structure is visualized (Fig. 5), see Santiago et al. (1998) for details.

Numerical 3D steady and unsteady single-phase Navier–Stokes solutions for the Y1-channel have been computed for the range of experimentally tested flow rates, corresponding to the interval 0.01 m/s < \( V \) < 0.3 m/s. The results confirm that the 3D numerical solutions give a very good representation of the observed flow fields. The numerical velocity distributions and the corresponding pathlines are consistent with the PIV measurements and visualizations, offering a better insights of the vortical structures within the closed branch (Fig. 5).

Validation of the experimental data by numerical simulations was also obtained for weakly elastic polymer solutions (polyacrylamide...
solutions in water – PS sample, Fig. 6), whose viscosity functions are well described by the Carreau–Yasuda model,

\[
\frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = \left[1 + \left(\frac{\dot{\gamma}}{\gamma_0}\right)^\mu\right]^\frac{\nu-1}{\nu}
\]

see Fig. 7.

The above experimental tests confirm that numerical procedures based on the solvers implemented in the FLUENT code are giving correct representations of real complex flows in micro-bifurcations and a fair description of the interface shape between immiscible fluids.

3. Simulations results

The main aim of the paper is to model the flow dynamics in the vicinity of the non-stationary interface between immiscible fluids, Fig. 7.

The simulations are 2D and represent the time evolution of the streamlines, vortical structures and interface position in the Y2-channel.

**Fig. 9.** Dynamics of the interface between immiscible Newtonian fluids: surface tension influence. The two liquids have properties of water; symmetric entrance velocities – \(V = 1\) m/s. The simulations are 2D and represent the time evolution of the streamlines, vortical structures and interface position in the Y2-channel.

**Fig. 10.** Details of the vortex within the closed branch (corresponding to the last pictures from Fig. 8).
liquids. In all simulations the interfacial surface tension is maintained constant at $\sigma = 0.05 \text{ Pa m}$, and the conditions are isothermal and isochoric. The flow is unsteady and the Newtonian liquids are assigned a constant viscosity of $\eta = 1 \text{ mPa s}$. Zero velocity on the channels wall was imposed as boundary condition, but no additional constraints for the VOF solution of the interface are imposed (i.e. contact angle, wall adherence). The influence of shear thinning behaviour is investigated using the Carreau–Yasuda model (1). Here, all simulations are performed with the non-stationary 3D-VOF solver. The phase-B (Newtonian fluid) is the primary one (i.e. it “fills” initially the flow geometry) and the secondary phase-A (Newtonian or shear thinning fluid) enters the channel. The time step between two iterations is $10^{-5} \text{ s}$ and the absolute convergence of solution is $10^{-10}$ within each step. At starting computation time ($t = 0.0 \text{ ms}$) the phase-A enters the microchannel with constant velocity $V$, the entrance being located $7 \text{ mm}$ upstream the junction (the boundary condition at the exit of the open branch is constant pressure). The computations (performed on a PC with eight parallel processors) were limited to at the first $100 \text{ ms}$, which was sufficient to reach almost the steady-state solutions in the presence of interfacial surface tension. In Figs. 8 and 9 the influences of surface tension on the evolution of the interface between two Newtonian fluids with identical properties are shown. Fig. 10 shows the vortices formed in the closed branch of the Y1-channel geometry, in the absence and in the presence of surface tension.

The simulations bring out the major influence of surface tension on the flow pattern. Without surface tension the secondary fluid gradually replaces the primary phase from the bifurcations. In the case of Y1-channel the fluid A penetrates into the close branch and forms a well defined 3D-vortical structure. The presence of surface tension limits the contact between phases, the two immiscible fluids always being separated by a well defined interface, the close branch remaining filled with the primary fluid. The computed dynamics of vortical structures in the vicinity of the interface with surface tension included is shown in Fig. 11. In this case, the vortices follow the interface and generate a significant velocity component ($v_z$) normal to the main flow direction (i.e. $z$-direction), in comparison to the case without surface tension, see Fig. 12a. In this case, the interface is passing, the intensity of $v_z$ is decreasing and the motion becomes almost 2D along the main flow direction (Fig. 12b).

The vortices in the secondary phase decrease with decreasing the Reynolds number (Fig. 12a) and they completely disappear for $Re < 1$. This is the case for simulations performed with PS (phase-A) and water (phase-B), Fig. 13, where the viscosity of the PS sample is $\eta > 30 \text{ mPa s}$ (for the corresponding strain rates, $10 \text{ s}^{-1} < \dot{\gamma} < 100 \text{ s}^{-1}$, see Fig. 6).

In the presence of surface tension, the influence of some other material parameters on the local hydrodynamics have been also investigated. One parameter under study was the $n$-shear thinning index associated to the viscosity function (1) of fluid A. Simulations did not show any remarkable effects of $n$-magnitude on the interface shape, if the ratio between the zero and infinite viscosities is maintained constant. The shear thinning influence is limited to the distribution of $v_z$; since the viscosity is not homogeneous, the magnitude of $v_z$ increases at the wall, where the strain rate and the local Reynolds number is higher than in the centre of the channel, see Fig. 12a.

4. Conclusions

The main goal of the present study was to investigate and model the formation of vortices in the vicinity of the interface between two immiscible fluids, during the dislocation of one liquid by another liquid in a micro-branching geometry. The work has focused on the 3D numerical simulations of the unsteady flow patterns in a micro-bifurcation generated by the travelling of a separation surface.

The VOF subroutine from the commercial FLUENT code (unsteady laminar and isochoric solvers) was first tested and validated against several experiments, performed with Newtonian and weakly elastic viscous fluids, on two Y-channel geometries with square cross-sections. In all cases accurate representations of the experimental flow spectra and interface shape are obtained, which confirms that numerical computations provide an accurate representation of the real flow patterns within the channel.

The presence of interfacial surface tension determines the shape of a clearly defined separation surface between the contact phases. In its neighbourhood, transitory 3D-vortices are formed, a phenomenon which determines the local mixing within each phase, even at moderate and low Reynolds numbers. This unsteady
complex hydrodynamics develops rapidly in a microchannel, but it is well captured and described in our numerical simulations.

Even if the present studies are limited to only the flows of immiscible Newtonian and generalized Newtonian liquids, the results offer a fair description of the unsteady patterns developed in micro-bifurcations and their interdependence on the evolution of the interface shape. The modelling of the 3D-vortical structures during the displacement of the primary phase from the channel contains valuable information in developing novel lab-on-a-chip applications, especially if a diffusion process between phases follows that dynamical process.

We believe that interfacial surface tension and normal stresses (together with no-slip conditions at the walls) might be the major factor which influences the mixing process of complex fluids in microchannels at low Reynolds numbers. This assertion is now being tested experimentally with work in progress being focused on obtaining PIV measurements of the micro-flows velocity distributions in the presence of the interface between a Newtonian oil and polymer solutions with different concentrations of polyacrylamide in water.

Acknowledgements

The present work was founded by the Romanian National Research Council – CNCSIS Grants A102 (2006–2008) and BD 73 (2008–2010). The authors acknowledge the supports of Prof. Thorsten Wohland (NU Singapore), in obtaining the Y2-channel confocal microscopic flow visualizations, Dr. ing. Tiberiu Barbat, for his advice on using the CFD techniques.

References


