Unsteady mixing of binary liquid mixtures with composition-dependent viscosity

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H I G H L I G H T S

- Direct Numerical Simulations of laminar mixing of five non-ideal liquid mixtures in a T junction.
- Determination of vortex, engulfment, unsteady and chaotic regimes for each mixture.
- All regimes strongly affected by the dependence of viscosity non composition.
- Time-periodic regimes with Strouhal number = 0.1–0.3 observed for all mixtures.
- Degree of mixing largely enhanced in the time-periodic regime.

A R T I C L E   I N F O

A B S T R A C T

The laminar mixing of two miscible liquids in a T junction is investigated through Direct Numerical Simulations. In particular the liquids have the same density and the same viscosity, but the viscosity of the mixture strongly depends on composition and can be up to three times larger or smaller than that of the pure liquids; this is aimed at mimicking the behavior of many aqueous and organic mixtures of interest in the pharmaceuticals and fine chemistry industry. The mixing process is found to be largely affected by the dependence of viscosity on composition. When the viscosity of the mixture is larger than that of the pure components, a viscous layer forms at the confluence of the inlet flows, which tends to keep the two streams separated, resulting in a shift of the engulfment regime towards larger Reynolds numbers. On the contrary, when the viscosity of the mixture is lower than that of the pure components, an acceleration of the fluid at the center of the mixing channel takes place, leading to an enhancement of the degree of mixing. At larger Reynolds number, unsteady, time periodic flow regimes, able to strongly increase the degree of mixing, are established for all types of mixtures. Although fluid oscillations are all characterized by typical Strouhal numbers lying in the range 0.1–0.3, the onset of the unsteady regimes, together with the corresponding vortical structures, strongly depends on the fluid rheology.

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1. Introduction

A fast and efficient mixing at the microscale is required in many processes ranging from miniature fuel cells to molecular diagnostics. Among passive micro-mixers, where mixing is promoted without the help of any external power source (Kumar et al., 2011), one of the simplest designs is a T shape, in which the inlets join the main channel with T-shaped branches. This type of mixer is also suitable for carrying out fundamental studies, as it is often the junction element in more complex micro-systems. The efficiency of T-shaped micro-mixers for liquid mixing has been largely investigated in the literature and different flow regimes have been

identified, both numerically (Bothe et al., 2006; Dreher et al., 2009; Fani et al., 2014; Andreussi et al., 2015) and experimentally, using water as working fluid (see μ-LIF and μ-PIV experiments by Hoffmann et al. (2006), and the flow visualizations by Engler et al. (2004)). In particular, it was shown that the inlet streams remain separated up to a critical Reynolds number, $Re = 140$, corresponding to the transition from a vortex flow regime, with a double mirror symmetry, to an engulfment flow regime, with a point central symmetry, leading to a large increase of the degree of mixing. Then, a few investigations have indicated that by further increasing Re, the flow becomes first unsteady and time-periodic at $Re = 240$, and eventually chaotic at $Re = 500$ (Dreher et al., 2009; Thomas et al., 2010; Thomas and Ameel, 2010; Kockmann and Roberge, 2011; Minakov et al., 2012, 2013). These results were explained by Fani et al. (2014) and extended by Andreussi et al.

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(2015) who investigated the effect of geometry on the flow regimes. In particular, in agreement with Thomas et al. (2010) and Thomas and Ameel (2010), they observed an unexpected transition from an unsteady asymmetric regime, i.e. presenting vortical structures with two main legs in the mixing channel, to an unsteady symmetric regime, i.e. characterized by four main legs with a double-mirror symmetry, which may dampen the mixing process.

The practical use of micro-devices involves mixing of two different fluids, often organic ones, as it is the case in pharmaceutical and fine chemistry industry. Hence the validity of the aforementioned results, obtained by feeding the same fluid, i.e. water, from both inlets, should be checked when using practical fluids and mixtures, that often show an non-ideal behavior, i.e. the viscosity is not a linear function of the composition. In a recent work (Galletti et al., 2015), we studied the engulfment process in a T-shaped micro-device for a model binary mixture, composed of two fluids having the same density and the same viscosity, yet presenting a strong fluidity of mixing effect, i.e., the viscosity of the mixture is a function of its composition. In the case of a positive fluidity of mixing, when the fluid mixture has a smaller viscosity than that of its pure components, the onset of the engulfment regime is accompanied by a sharp increase of the degree of mixing, with the critical Re decreasing as the fluidity of mixing increases. The contrary occurs when the fluid mixture has a larger viscosity than that of its pure components. This result is in agreement with previous findings by Orsi et al. (2013a), who compared the case where the two inlet fluids are both water with that where one inlet fluid is water and the other is ethanol, finding that the vortex-engulfment flow transition occurs at larger Re numbers for the water-ethanol case than for water-water case (i.e., $Re = 230$ vs. $Re = 140$). The reason of this result is that a water-ethanol mixture has a viscosity that is up to three times larger than that of water, so that at the confluence of the T-mixer, the two streams are kept separated from each other by a viscous interfacial layer that hampers vortex formation and retards mixing (Orsi et al., 2013b). However, it was also observed that at low Re the presence of viscosity gradients ensures some degree of mixing, albeit small.

In the present work, we want to corroborate previous findings and better investigate the dependence of the mixing process on the shape of the viscosity-composition diagram $\mu = \mu(\phi)$. In addition, we want to investigate how the mixing process takes place at larger Re, when the flow regime becomes first time periodic and then chaotic. Hence, we intend to characterize the transitions between different types of steady and unsteady regimes and how they depend on the mixture rheology; this will provide a large database, encompassing all cases that might be of interest in microfluidics.

2. Problem description

2.1. Constitutive relations

In order to investigate the effect of a composition-dependent viscosity on mixing, we assume that the two fluids composing the mixture have the same density, ρ, and the same viscosity, μ_0, and that, upon mixing, density remains constant, while the mixture is a Newtonian fluid, with a composition-dependent viscosity. The following types of constitutive relations are considered:

B base case, for which $\mu(\phi) = \mu_0$;
N1 negative fluidity of mixing, with a symmetric quadratic composition diagram,

$$\mu(\phi) = \mu_0[1 + K_1\phi(1 - \phi)]; \quad K_1 = (\alpha - 1)/\beta^2. \tag{1}$$

N2 negative fluidity of mixing, with a symmetric and sharp composition diagram,

$$\mu(\phi) = \mu_0\left[1 + K_2\phi^3(1 - \phi)^3\right]; \quad K_2 = \beta^{-7}. \tag{2}$$

S sinusoidal viscosity composition diagram,

$$\mu(\phi) = \mu_0[1 + K_3\sin(2\pi\phi)]; \quad K_3 = \frac{1}{2}. \tag{3}$$

P1 positive fluidity of mixing, with a symmetric quadratic composition diagram,

$$\mu(\phi) = \mu_0[1 - K_4\phi(1 - \phi)]; \quad K_4 = (\alpha - 1)/(\alpha\beta^2). \tag{4}$$

In the N1- and N2-type mixtures, viscosity reaches its maximum value, $\mu_{max} = \alpha\mu_{min} = \alpha\mu_0 = 3\mu_0$, at $\phi = \beta = \frac{1}{2}$, that is for a 50–50% composition; in the P1-type mixture, viscosity presents a minimum value, $\mu_{min} = \mu_0/\alpha$, at $\phi = \beta = \frac{1}{2}$, that is for a 50–50% mixture; in the S-type mixture, viscosity has a maximum, $\mu_{max} = \frac{3}{2}\mu_0$, at $\phi = \frac{1}{4}$ and a minimum, $\mu_{min} = \frac{1}{2}\mu_0$, at $\phi = \frac{3}{4}$. The viscosity-composition diagram is reported in Fig. 1 for each mixture type. Note that, in all cases, $\mu_{max}/\mu_{min} = 3$; this choice was made to emulate the behavior of one of the most common mixtures, namely water-ethanol, whose maximum viscosity is almost three times that of pure water (Simmonds, 1919).

Examples of N-type mixtures are many aqueous solutions of organic solvents, such as ethanol, acetone, methanol, propanol and acetic acid (Dizechi and Marschall, 1982; Laliberté, 2007); ethanol/methylacetate and ethanol/benzene are examples of P-type mixtures, while benzene/methanol and toluene/methanol are examples of S-type mixtures (see Fig. 2 of González et al., 2007, and references therein; Figs. 1–4 of Lei and Hou, 1999).

The main idea behind these simulations is to investigate the dependence of the degree of mixing on the viscosity gradients. In fact, during the mixing process of a fluid mixture, convective flows are induced by viscosity gradients, due to the non-homogeneous distributions of composition and temperature. Accordingly we expect that, for example, comparing the N1 with the N2 case, as in the latter the viscosity gradient reaches a larger value than in the former case, the convective flux will be larger, therefore enhancing the mixing process.

2.2. T-mixer geometry

The geometry of the problem consists of the T-shaped micromixer illustrated in Fig. 2, where the coordinate y denotes the distance from the confluence of the inlet streams. Here, the inlet channels have a square cross section, with $W_i = H$, while the mixing channel presents a 2 : 1 aspect ratio, i.e. $W = 2H$, so that the
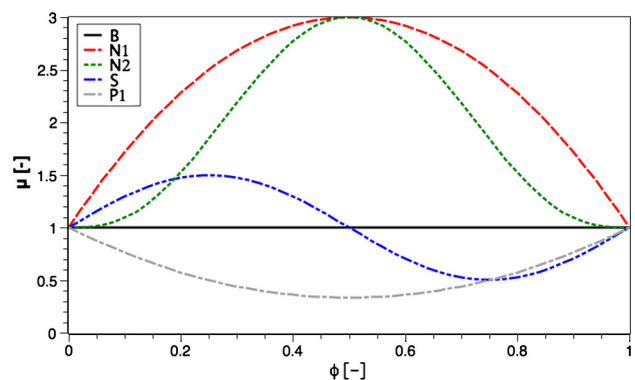

Fig. 1. Viscosity as a function of composition for all investigated mixtures.