Influence of Viscosity Ratio on Droplets Formation in a Chaotic Advection Flow

Charbel Habchi∗  Sofiane Ouarets†  Thierry Lemenand‡
Dominique Della Valle**  Jérôme Bellettre††  Hassan Peerhossaini‡‡

∗LTN, Ecole Polytechnique, University of Nantes, charbel.habchi@univ-nantes.fr
†LTN, Ecole Polytechnique, University of Nantes, sofiane.ouarets@univ-nantes.fr
‡LTN, Ecole Polytechnique, University of Nantes, thierry.lemenand@univ-nantes.fr
**LTN, Ecole Polytechnique, University of Nantes, dominique.dellavalle@univ-nantes.fr
††LTN, Ecole Polytechnique, University of Nantes, jerome.bellettre@univ-nantes.fr
‡‡LTN, Ecole Polytechnique, University of Nantes, hassan.peerhossaini@univ-nantes.fr

ISSN 1542-6580
Copyright ©2009 The Berkeley Electronic Press. All rights reserved.
Influence of Viscosity Ratio on Droplets Formation in a Chaotic Advection Flow

Charbel Habchi, Sofiane Ouarets, Thierry Lemenand, Dominique Della Valle, Jérôme Bellettre, and Hassan Peerhossaini

Abstract

The efficiency of liquid/liquid dispersion is an important sake in numerous sectors, such as the chemical, food, cosmetic and environmental industries. In the present study, dispersion is achieved in an open-loop reactor consisting of simple curved pipes, either helically coiled or chaotically twisted. In both configurations, we investigate the drop breakup process of two immiscible fluids (W/O) and especially investigate the effect of the continuous phase viscosity, which is varied by the addition of different fractions of butanol in the native sunflower oil. The global Reynolds numbers vary between 40 and 240, so that the flow remains laminar while the Dean roll-cells in the bends develop significantly. Different fractions of butanol are added to the oil in each case to examine the influence of the continuous phase viscosity on the drop size distribution of the dispersed phase (water). When the butanol fraction is decreased, the dispersion process is intensified and smaller drops are created. The Sauter mean diameters obtained in the chaotic twisted pipe are compared with those in a helically coiled pipe flow. The results show that chaotic advection intensifies the droplet breakup until there is a 25% reduction in droplet size.

KEYWORDS: drop-breakup diagram, chaotic advection, emulsification, multifunctional heat exchanger-reactor

*This work was partially supported by ADEME. Authors would like to acknowledge the continuous support of Dr. Gwenaël Guyonvarch, the Ile de France Regional Director of ADEME and also Dr. Cédric Garnier for monitoring this grant. C. Habchi is supported jointly by ADEME and CNRS. Please send correspondence to H. Peerhossaini: Phone: + (33)-240 683 124, Fax: + (33)-240 683 141, e-mail address: hassan.peerhossaini@univ-nantes.fr.
1. INTRODUCTION

The mixing of immiscible fluids in industrial processes, for phase dispersion or emulsification, is a complex issue, since the breakup mechanisms are difficult to understand and quantify. One phase is dispersed into small droplets of various diameters. The drop size distribution is decisive for the final product properties, for instance emulsion texture and stability in a food product or the result of a chemical reaction at the interface. The dispersion process may also be aimed at producing efficiently emulsified fuel for combustion enhancement (Senthil Kumar et al., 2005, 2006; Tarlet et al., 2008). We report on a special mixing process achieved in an open-loop reactor consisting of simple curved pipe segments in both helically coiled and chaotic twisted pipe configurations.

In the present study, the effect of the continuous phase viscosity on droplet sizes is investigated in order to address the practical issue of viscosity optimization in emulsification. An aqueous phase (water) is dispersed in a continuous oily phase (commercial sunflower oil), the viscosity of which can be lowered by adding butanol without significant effect on the interfacial tension. The global effect of the viscosity reduction results in competitive effects that we call “positive” if they work toward a smaller droplet size, and otherwise “negative”. These effects are:
- reduction of the external stress, and hence weakening of the external forces able to deform and split the drops (negative effect),
- increase in Reynolds number and in Dean number, leading to intensification of the secondary flow and of the associated strain rates (positive effect),
- lowering of the critical capillary number that governs the breakup in laminar flow, in the viscosity range of interest in experiments (positive effect).

The effect of viscosity cannot be determined directly from the experimental results. In this work we attempt to interpret the experimental results through the theoretical insights provided by Taylor-Grace laminar breakup analysis (1953, 1982), and knowledge of the kinematic field in Dean flow provided by the asymptotic solutions of Dean (1928). The trends are well confirmed except for a lack of accuracy in the predictions, which can be attributed to flow complexity.

The forces due to the continuous-flow velocity gradients in the main flow create deformation strains. Grace (1982) underlines the existence of two strain modes, shear strain and extensional strain, whose effects on the breakup mechanism are quite different. The internal viscous forces depend on the ratio $p$ of the viscosities of the dispersed and continuous phases:

$$p = \frac{\mu_d}{\mu_c}$$

(1)
where $\mu_c$ and $\mu_d$ are respective continuous and dispersed phase viscosities.

In order to characterize the relative effect of the different forces, a dimensionless parameter $Ca$, called capillary number, is defined:

$$Ca = \frac{\text{Viscous forces}}{\text{Capillary forces}} = \frac{d_{\text{drop}} \tau}{2\sigma}$$

(2)

where $d_{\text{drop}}$ is the drop diameter, $\sigma$ the interfacial tension, and $\tau$ the viscous stress.

Grace (1982) considered that drop breakup occurs when the capillary number exceeds some critical value that depends upon the viscosity ratio $p$, which reflects the drop’s internal viscous resistance.

Previous work by the present authors (Habchi et al., 2009) studied the dispersion in both helically coiled and chaotic twisted pipe flows. It was shown that an increase in flow rate decreases drop size, and that chaotic advection flow intensifies the emulsification process and provides smaller and more homogeneously dispersed droplets. Both Eulerian and Lagrangian analytical studies showed that chaotic advection causes fluid particles to visit regions of high shear and elongation rates, thus intensifying emulsification. However, this study was limited to low Reynolds numbers (not above 70). Therefore, in order to increase the Reynolds numbers range, we decreased the continuous phase viscosity here by adding butanol fraction, eliminating the need to increase the flow rate. In this paper we investigate the effect of the continuous phase viscosity on droplet breakup in both helically coiled and chaotic twisted pipe flows. The experiments carried out in both configurations allow comparison between the two cases. It appears that the latter, which has already been shown to improve heat transfer (Mokrani et al., 1998), also provides better performance for liquid/liquid dispersion.

This paper is organized as follows: in section 2, experimental apparatus and measurement methods are described. Results and discussion are given in section 3, and section 4 is devoted to conclusions.

2. EXPERIMENTAL APPARATUS AND METHODS

2.1 Hydraulic Loop and test sections

A schematic diagram of the hydraulic loop used in the experiments appears in Figure 1. Experiments were carried out for two-phase flow using immiscible fluids. The experimental setup consists of two similar loops. Each working fluid is contained in a tank. The oil is pumped by a centrifugal pump, and the water is
supplied by a constant-level feed tank connected to the water supply. Flow rates are controlled by valves and measured with two flowmeters. The same experimental setup was used by Lemenand et al. (2003, 2005), for turbulent dispersion and Habchi et al. (2009) for laminar and chaotic advection flows. Water is injected at the test section inlet by an injection needle of inside diameter 2 mm, designed not to disturb the main flow and not to create additional breakup of the dispersed phase. For accurate evaluation of breakup performance, the volume fraction of the dispersed aqueous phase is very low (about 1%) so that coalescence phenomena are negligible. Water injection flow rate is small enough to prevent the coalescence that can occur in the visualization box, and to minimize the main flow disturbance. Bigger drop diameters appear when smaller droplets enter in collision.

![Figure 1: Schematic diagram of the hydrodynamic loop for droplet formation](image)

The test section, composed of a succession of 90° bends, is designed to be arranged in two configurations: helically coiled pipe and chaotic twisted pipe, where each bend is rotated by 90° with respect to the preceding one. In laminar flow through curved pipes, centrifugal forces create a secondary flow in the radial direction consisting of a pair of counter-rotating roll-cells known as Dean roll-cells (Dean 1927, 1928). Chaotic advection is achieved by alternating the bend orientation of ± 90°. This geometrical perturbation greatly increases the heat and mass transfer (Jones et al., 1989; Acharya et al., 1992; Jones and Young, 1994; Peerhossaini et al., 1993; Castelain et al., 2000; Mokrani et al., 1998; Lemenand and Peerhossaini, 2002; Habchi et al., 2009). The test section dimensions are summarized in Table 1.
Table 1: Test section dimensions

<table>
<thead>
<tr>
<th></th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radius of circular pipe</td>
<td>4.00 mm</td>
</tr>
<tr>
<td>Bend curvature radius</td>
<td>44.00 mm</td>
</tr>
<tr>
<td>Curvature angle in bend plane</td>
<td>$\pi / 2$ rad</td>
</tr>
<tr>
<td>Number of bends</td>
<td>25</td>
</tr>
<tr>
<td>Total curved length</td>
<td>1.80 m</td>
</tr>
<tr>
<td>Total straight length between bends</td>
<td>0.20 m</td>
</tr>
<tr>
<td>Total length</td>
<td>2.00 m</td>
</tr>
</tbody>
</table>

2.2 Working Fluids

Experiments were carried out for a two-phase flow using immiscible fluids. Working fluids are vegetable oil for the continuous phase and water for the dispersed phase. Different mass fractions of butanol are added to the continuous phase in order to modify the continuous phase viscosity $\mu_c$. Physical properties of the fluids and measurement methods are given in Table 2.

Table 2: Characteristics of working fluids

<table>
<thead>
<tr>
<th>Properties (at 298 K)</th>
<th>Values</th>
<th>Measurement methods</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{\text{oil+25% butanol-water}}$</td>
<td>0.036 ± 0.003 N/m</td>
<td>Krüss™ tensiometer (K12) by ring method</td>
</tr>
<tr>
<td>$\rho_c$</td>
<td>889 Kg/m³</td>
<td>Data Technical™</td>
</tr>
<tr>
<td>$\mu_c$</td>
<td>0.052 Pa s</td>
<td>Mettler™ RM180 rheometer</td>
</tr>
<tr>
<td>$\rho_d$</td>
<td>998 Kg/m³</td>
<td>Data Technical™</td>
</tr>
<tr>
<td>$\mu_d$</td>
<td>0.001 Pa.s</td>
<td>Mettler™ RM180 rheometer</td>
</tr>
</tbody>
</table>

Oil viscosity is sensitive to temperature variation; this effect is taken into consideration. The oil temperature is controlled by a chromel/alumel (type K) thermocouple in the oil admission circuit. Studies show that a 25% addition of butanol decreases halves the continuous phase viscosity; increasing the Reynolds number while conserving the flow rate:
\[
\mu_{oil} = \mu_{oil, T_0} \exp \left[ \frac{E}{R} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right]
\]  

(3)

where \( E \) is activation energy, \( \mu_{oil, T_0} \) is viscosity at a reference temperature \( T_0 \), and \( R = 8.314 \text{ J K}^{-1}\text{mol}^{-1} \).

The variation of the continuous phase viscosity with temperature and butanol fraction is graphed in Figure 2. As shown, an addition of 25% of butanol halves the continuous phase viscosity, increasing the Reynolds number while conserving the flow rate. This addition decreases the deformation strain rates in the main flow. Moreover, the continuous phase viscosities decrease when temperature is increased, with the same variation, whatever the butanol fraction.

![Figure 2: Continuous phase viscosity](image)

The consequence of varying the continuous phase viscosity is detailed in section 3. Measured viscosities are fitted with continuous lines in Figure 2 by using Arrhenius’ law, as given by equation (3). The activation energies \( E \) correlated from measurements are given in Table 3.
Table 3: Activation energy for different butanol fractions

<table>
<thead>
<tr>
<th>Butanol to oil fraction</th>
<th>0%</th>
<th>5%</th>
<th>10%</th>
<th>15%</th>
<th>20%</th>
<th>25%</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E$ (kJ)</td>
<td>26.15</td>
<td>23.48</td>
<td>25.24</td>
<td>23.79</td>
<td>22.07</td>
<td>22.82</td>
</tr>
<tr>
<td>Fitting accuracy (%)</td>
<td>99.22</td>
<td>99.53</td>
<td>99.09</td>
<td>99.34</td>
<td>99.45</td>
<td>99.05</td>
</tr>
</tbody>
</table>

2.3 Droplet Diameters Measurement

Emulsion pictures are taken by a high-frequency digital Canon™ camera whose optical axis is perpendicular to the plane of the rectangular visualization window. The visualization system, a parallelepiped box with 2D diffuser at the entrance and 2D nozzle at the exit, is placed directly at the test section outlet (shown in Figure 1). The visualization system is designed to minimize flow disturbances and to maintain the same deformation strain levels as in the test section. For each operating condition, the sequence of independent images recorded constitutes our statistical sample of the drops. Drop diameters are measured from the recorded images. Samples of diameter distribution should be independent of the population (number of droplets). Therefore, we measured at least 400 drops in each run.

Reproducibility testing was carried out to check the effect of a new operator and new trial on the measured diameters of the final size distribution. The maximum standard deviation based on Sauter mean diameter was found to be less than 7%, implying that the measurements are reproducible.

3. RESULTS AND DISCUSSIONS

3.1 Experimental plan

The objective of the present study is to investigate the effect of the continuous phase viscosity on the final drop size distribution. The viscosity variation depends on several parameters such as temperature and the addition of surfactants. The interfacial tension is modified due to the addition of butanol: as shown in Table 2, for 25% butanol fraction in the oil, the interfacial tension changes by about 30%. However, in the present butanol range [0% - 12.5%], the interfacial tension variation is less than 4% and can be neglected. As a consequence, the flow rate and the continuous phase viscosity are the only parameter contributing to the emulsification process.

The drop size distribution in the flow is characterized by the Sauter mean diameter. This diameter, given in equation (4), represents the characteristic drop diameter as:
\[
d_{32} = \frac{\int \ell^3 f(\ell) d\ell}{\int \ell^2 f(\ell) d\ell}
\]

(4)

here \(f(\ell)\) is the distribution function representing the proportion of drops having a given diameter \(\ell\) in the observed emulsion.

3.2 Experimental data

The measured Sauter mean diameters in the helically coiled and chaotic twisted pipe are represented in Figure 3 as functions of the continuous phase viscosity \(\mu_c\) for Reynolds number 60. The Sauter mean diameter \(d_{32}\) decreases with \(\mu_c\), showing that the emulsification process is greatly enhanced in both regular and chaotic flows by the butanol addition. The physical interpretation of this phenomenon is explained below using Grace (1984) theory.

Figure 3: Sauter mean diameter as a function of viscosity ratio for fixed Reynolds number \(Re = 60\)
3.3 Interpretation by Grace-Taylor breakup diagram

To interpret the physical effect of the continuous phase viscosity on drop breakup, a Grace diagram is adopted by recalculating the critical capillary number as:

\[ Ca_{\text{critical}} = \frac{\mu_s \dot{\gamma}_{\text{effective}} d_{\text{max}}}{2\sigma} \]  

(5)

\( d_{\text{max}} \) is the droplet’s maximum diameter.

In the range of Reynolds number studied here, the effective deformation strain rate \( \dot{\gamma}_{\text{effective}} \) is considered equal to the maximum shear rate in a helically coiled pipe flow, \( \dot{\gamma}_{\text{Dean}} \), which has been computed by Habchi et al. (2009) as:

\[ \dot{\gamma}_{\text{Dean}} = 4 \frac{W}{a} \left[ 1 + \frac{1}{150} \left( \frac{a}{2R} \frac{Re}{\text{Re}} \right)^2 \right]^{0.5} \]  

(6)

where \( a \) is the radius of circular duct, \( R \) the radius of curvature, and \( W \) the flow mean velocity.

However Habchi et al. (2009) gives only an order of magnitude for \( Ca_{\text{critical}} \) in a small range of viscosity ratio \( p \). The theory is established in pure flows (simple shear or simple elongation) that allow homogeneous deformation rates. In the present study, the flows are more complex, and the heterogeneity distribution of the effective strain rates adopted here limits the accuracy in calculating \( Ca_{\text{critical}} \). Therefore; \( \dot{\gamma}_{\text{effective}} \) is merely an indicative value for the deformation strain rates, and uncertainty appears when calculating \( Ca_{\text{critical}} \). In Figure 4, the critical capillary numbers are represented for both mixers; they merge to a master curve in the Grace (1982) diagram.

Figure 4 shows that the present results for \( Ca_{\text{critical}} \) are located in the zone where \( p \) should increase to transmit the deformation forces more effectively from the continuous to dispersed phase. This explains the intensification of the emulsification process, due to the decrease in continuous phase viscosity, which is caused here by the addition of butanol.
To recapitulate, when the continuous phase viscosity decreases, three parameters are modified:
- The deformation rate increases
- The Reynolds number increases, thus intensifying the strain rate in the Dean flow $\gamma_{Dean}$ given in equation (6)
- The viscosity ratio $p$ tends to the optimum value of 1

Therefore, the emulsification process is highly intensified by decreasing the continuous phase viscosity.

![Figure 4: Drop breakup diagram](image)

**3.4 Chaotic advection effect**

In the twisted pipe configuration, Dean roll-cells are reoriented after each bend, and stretching and folding are more intense than in the helically coiled pipe. This geometrical perturbation creates chaotic advection trajectories whereby the dispersed-phase fluid visits the whole mixer cross section, passing by the maximum
values of shear and elongation rates. Droplet breakup is then enhanced and the final drop diameters are smaller in twisted-pipe Dean flow than in regular Dean flow (i.e., in the helically coiled mixer). This effect is shown in Figure 5, which compares the two mixers for different Reynolds numbers.

Figure 5: Sauter diameters: comparison between helically coiled and chaotic twisted pipe flows

Considering the relative improvement over regular laminar flows (Figure 6), we see droplet diameters between 17 to 31% lower in chaotic advection twisted pipe flow than in helically coiled tube flow. Mokrani et al. (1998) have shown that chaotic advection in twisted pipe flow improves heat transfer of about 30% above the helically coiled pipe.
4. CONCLUSIONS

The reduction of the continuous phase viscosity intensifies the emulsification process in the two geometries considered, helically coiled and chaotic twisted pipe flows: the Dean flow effect is decisive in this process. When 25% butanol is added to the oil, the continuous phase viscosity is halved, so that it was possible to increase the Reynolds number without needing to increase the flow rate. The chaotic advection strongly enhances mass transfer and gives raise to smaller droplet size distribution (by about 25% in average) than in regular Dean flow. Theory provides useful support for process design and future improvements or optimization, for example in fuel production.
NOMENCLATURE

\[ a \quad \text{radius of circular duct (m)} \]
\[ Ca \quad \text{capillary number (–)} \]
\[ d_{\text{drop}} \quad \text{drop diameter (m)} \]
\[ d_{\text{max}} \quad \text{maximum diameter in size distribution (m)} \]
\[ d_{32} \quad \text{Sauter mean diameter (m)} \]
\[ E \quad \text{activation energy (kJ)} \]
\[ \frac{p = \mu_d / \mu_c}{R} \quad \text{viscosity ratio (–)} \]
\[ R \quad \text{radius of curvature (m)} \]
\[ Re \quad \text{Reynolds number, } Re = \frac{2Wa}{\nu} \quad (–) \]
\[ T \quad \text{temperature (K)} \]
\[ W \quad \text{flow mean velocity (m s}^{-1}) \]

Greek letters
\[ \mu \quad \text{dynamic viscosity (Pa s)} \]
\[ \nu \quad \text{kinematic viscosity (m}^2 \text{s}^{-1}) \]
\[ \sigma \quad \text{interfacial tension (N m}^{-1}) \]
\[ \tau \quad \text{viscous stress (Pa)} \]
\[ \dot{\gamma}_{\text{effective}} \quad \text{effective shear rate (s}^{-1}) \]
\[ \dot{\gamma}_{\text{Dean}} \quad \text{computed shear rate in Dean flow (s}^{-1}) \]

Subscripts
\[ c \quad \text{continuous phase (oil)} \]
\[ d \quad \text{dispersed phase (water)} \]
REFERENCES


