# Co-current horizontal flow of a Newtonian and a non-Newtonian fluid in a microchannel

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**Abstract:** In this work, the flow of two immiscible liquids in a glass microchannel, *I.D.* = 580μm, was experimentally investigated. Various aqueous glycerol solutions containing xanthan gum were the non-Newtonian fluids, while kerosene was the Newtonian one. The flow rate of the non-Newtonian fluids varied from 50 to 200 μL/min, while the kerosene flow rate was kept constant. The two fluids were put in contact at a T-junction. Visual observations were made using a high speed *CCD* camera and data were collected by processing the corresponding video images. The flow pattern was slug flow irrespective of the fluid that initially filled the microchannel. The experimental results revealed that the length of the kerosene slugs decreases by increasing either the aqueous phase flow rate or its viscosity. Furthermore the non-Newtonian fluid results in smaller and more frequent slugs than the corresponding Newtonian one. Thus by rendering a fluid non-Newtonian the interfacial area increases and consequently the mass transport performance is enhanced. This observation is expected to aid to the optimal design of two-phase microreactors. More work is certainly needed to investigate the effect of all the design parameters on the characteristics of this kind of flow in microchannels.

**Keywords:** Two phase flow, microchannels, non-Newtonian, slug flow.

## 1. Introduction

Operations in microchannels have recently drawn worldwide interest due to their significant advantages. These devices, in comparison with conventional reactors, create higher surface to volume ratio, short transport path and in the same time they are relatively cheap and safe (Su et al., 2010; Boogar et al., 2013). Moreover, due to their small dimensions, the specific interfacial area of multiphase systems is increased, improving in this way, the mass and heat transfer rates between two immiscible phases (Dessimoz et al., 2008). In order to evaluate potentials and prospects of this new and promising technology in chemical engineering, a detailed knowledge of transport processes in microchannels is necessary (Su et al., 2010).

Microfluidic processes of liquid-liquid systems occur in a wide range of applications in chemical and petroleum engineering such as nitration, extraction and emulsification. Liquid–liquid two-phase flow patterns are formed when two immiscible fluids are brought in contact at the junction of a microchannel. Different patterns can be obtained depending not only on operational conditions but also on the properties of the fluids and geometrical characteristics of the microchannel (Jovanovic et al., 2011). The main flow patterns reported are slug, drop, annular and parallel flow depending on the competition between the interfacial tension and the inertia forces. In particular, ***slug flow*** is extensively studied due to its easily controllable hydrodynamics and its high mass transfer rate, which shows great potential for applications in various fields (Tsaoulidis et al, 2013). In this flow pattern, one liquid flows as a continuous phase while the other liquid (dispersed phase) flows in the form of slugs.

Due to their technological importance and applications in microreactors, a great number of studies have been conducted in liquid – liquid two-phase flow systems in microchannels (e.g. Talimi et al., 2012; Foroughi and Kawaji, 2011; Lovick and Angeli, 2004). For ***Newtonian*** systems, recently, Jovanovic et al. (2011) studied the effect of various operating conditions on the flow patterns and pressure drop and reported that the slug size depends on the slug velocity, the organic to aqueous flow rate ratio, the dynamic viscosity and the microchannel diameter.

Various investigators studied liquid-liquid two phase flow in T- and/or Y- microchannels. For example, Kashid and Agar (2007) reported that the capillary and Y-junction dimensions influence the slug size. Salim et al. (2008) studied the dependence of flow patterns and pressure drop on both the type of the fluid, which was initially filled the channel as well as the channel material, while Zhao et al. (2006) developed a flow pattern map divided into three zones depending on interfacial tension and inertia force. Dessimoz et al. (2008), who investigated the influence of fluid properties on the type of flow, developed a model based on the mean *Capillary* and *Reynolds* numbers for predicting the flow pattern. Finally, Tsaoulidis et al. (2013) have recently reported that the flow patterns of *Newtonian* liquid–liquid systems depend on the flow rate, the physical properties of both phases, the microchannel material and its wettability and the geometry of the mixing zone.

However, to the authors' best knowledge limited number of studies are available on liquid–liquid flow in small dimension conduits where one of the liquids exhibits *non-Newtonian* behavior (Boogar et al., 2013; Gu and Liow, 2011).

Our work is a ***preliminary*** report of a work concerning the experimental study of co-current horizontal flow of two immiscible fluids, namely a ***Newtonian*** liquid (organic phase) and a ***non-Newtonian*** *(shear thinning*) liquid (aqueous phase) in a circular glass microchannel. We aim to investigate the effect of the flow rate and the viscosity of the non-Newtonian fluid on the characteristics of this type of two-phase flow. In this initial stage the microchannel diameter and the flow rate and type of the organic phase were kept ***constant***.

## 2. Experimental Setup

The experiments were conducted in a cylindrical glass microchannel (*I.D.*=580μm, 12cm long). Two aqueous glycerol solutions containing xanthan gum were used as non-Newtonian (shear thinning) fluids, while kerosene was the Newtonian one. Xanthan gum is a polysaccharide that acts as rheology modifier and renders the fluid non-Newtonian. The compositions of all fluids used are presented in ***Table 1***. Three Newtonian fluids, *i.e.* aqueous glycerol solutions (*G1*, *G2* and *G3*) were also used for reference and comparison. The relevant properties of all fluids employed were carefully measured and are summarized in ***Table 2***.

***Table 1:*** *Composition of the working fluids.*

| Fluid Index | Working fluids |
|---|---|
| G1 | aqueous glycerol solution (30% w/w) |
| G1n | aqueous glycerol solution (30% w/w) + xanthan (0.035% w/v) |
| G2 | aqueous glycerol solution (50% w/w) |
| G2n | aqueous glycerol solution (50% w/w) + xanthan (0.035% w/v) |
| G3 | aqueous glycerol solution (70% w/w) |
| K | kerosene |

***Table 2:*** *Properties of the working fluids (T =20C).*

| Fluid Index | $\sigma_{A/K}$, (mN/m) | μ, cP | ρ, kg/m³ | φ, deg |
|---|---|---|---|---|
| G1 | 29 | 2.8 | 1070 | 52 |
| G1n | 24 | 3.2* | 1030 | 61 |
| G2 | 28 | 7.5 | 1125 | 58 |
| G2n | 25 | 8.3* | 1105 | 64 |
| G3 | 29 | 22 | 1180 | 48 |
| K | - | 2.2 | 800 | 23 |

* *the asymptotic value of viscosity .for the non-Newtonian liquids.*

The dynamic viscosity, $\mu$, of the Newtonian solutions was measured by a *KPG Cannon-Fenske* (*Shott*®) viscometer, while for the non-Newtonian solutions a magnetic bearing rheometer (*AR-G2 TA Instruments*®) was used. The interfacial tension, $\sigma$, and the contact angle, $\varphi$, of the solutions were measured using a *KSV CAM*® *200* tensiometer.

A schematic of the experimental setup is shown in ***Figure 1***. It consists of two sections: the mixing zone and the flow visualization section. Using two syringe pumps (*Aladdin-1000WPI*), the two immiscible fluids entered through the two inlet arms that meet at a T-junction. The flow visualization section comprises a high speed *CCD* camera connected to a Nikon (*Eclipse LV150*) microscope and a light source for the illumination of the test section. Image processing software (*AxioVision Zeiss*®) was used for extracting information from the acquired video images.

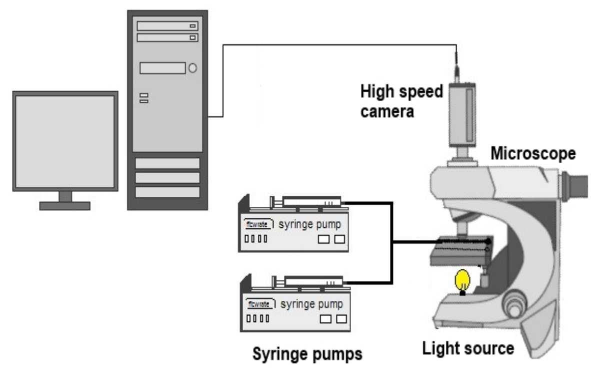


***Figure 1****: Experimental setup.*

Minute amounts of *Nigrosine* (a black dye) were diluted in the aqueous phase for better visualization of the flow. When steady state condition was established, the flow patterns were recorded at a frame rate of 600fps and at distance 10cm downstream the inlet junction. To eliminate image distortion caused by refraction due to the cylindrical walls, the microchannel was placed in a square cross-section *Plexiglas*® box filled with the aqueous solution of the corresponding experiment.

All runs were carried out at ambient temperature and pressure conditions (i.e. 20C and 1atm) and for a ***constant*** volumetric flow rate, namely 50μL/min, of the organic phase, which initially filled the microchannel, while the flow rate of the aqueous phase varied in the range of 50-200 μL/min.

From the recorded videos we extracted informations concerning the effect of the aqueous phase viscosity and flow rate on the flow patterns, the size and the frequency of slugs.

## 2. Experimental Results

***Flow patterns***

Apart from the experiments we conducted using non-Newtonian aqueous phases, experiments were also performed using the corresponding Newtonian solutions, i.e. without adding xanthan gum. This permits us to investigate the effect of the non-Newtonian behavior on the flow patterns as well as to compare our results with published data.

For our experimental conditions the organic phase forms slugs irrespective of the flow rate or the phase that initially filled the tube, an observation that is in accordance with what is reported in the literature (Boogar et al., 2013). Initially we observed that for the same flow rate, when the aqueous phase is Newtonian, the slugs have a symmetrical shape, i.e. their front and back ends are circular arcs (***Figure 2a***). The slugs have a "*bullet*" shape when the aqueous phase is non-Newtonian (***Figure 2b***).

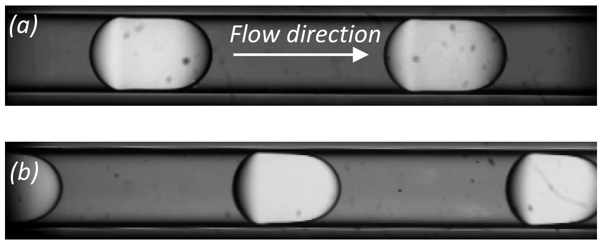


***Figure 2:*** *Typical slug shapes for: (a) Newtonian fluid G2 (b) non-Newtonian fluid G2n ($Q_A$=120 $\mu$L/min).*

This fact is better illustrated in ***Figure 3***, where the kerosene slugs are compared. It is obvious that, if all the other parameters are kept the same, the addition of a small amount of xanthan gum results in the formation of shorter "bullet" shape slugs (***Figure 3a*** and ***3b***). However it is interesting that Newtonian fluids with relatively high viscosity (*G3*, $\mu$=22cP) form also short "bullet" shape slugs (***Figure 3c***). This observation denotes that both the shape and the size of the slugs are dictated by the viscosity of the aqueous phase.

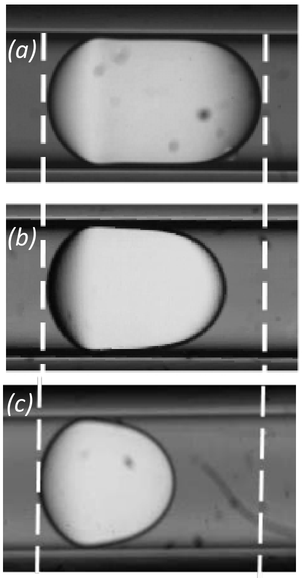


***Figure 3***: *Effect of type of liquid on slug characteristics for: (a) Newtonian fluid G2, (b) non-Newtonian fluid G2n, (c) Newtonian fluid G3.* ($Q_A$=120 μL/min).

The shape of slugs in microchannels is controlled by the force balance between interfacial tension, viscous and inertial forces (Zhao et al., 2006). It is obvious that while the interfacial tension tends to reduce the interfacial area, the drag force which depends on the continuous phase viscosity, affects the total force that acts on the interface towards the flow direction.

### *Effect of flow rate on the slug size*

The effect of flow rate on the slug size is illustrated in ***Figure 4*** for the *G2n.* Given that the kerosene flow rate is kept constant, the slug length depends only on the non-Newtonian liquid flow rate, i.e. by increasing the aqueous-phase flow rate the slug length decreases. Consequently the number of the produced slugs increases, i.e. the interfacial area increases, resulting in higher mass transfer rates. The same trend is observed during relevant experiments with Newtonian fluids performed in our Lab and is also confirmed by the work of Boogar et al. (2013).

From ***Figure 4*** we can also conclude that the shape of the slugs remains practically unaffected by the flow rate. This is also confirmed by the superposition of the images of slugs corresponding to different flow rates.

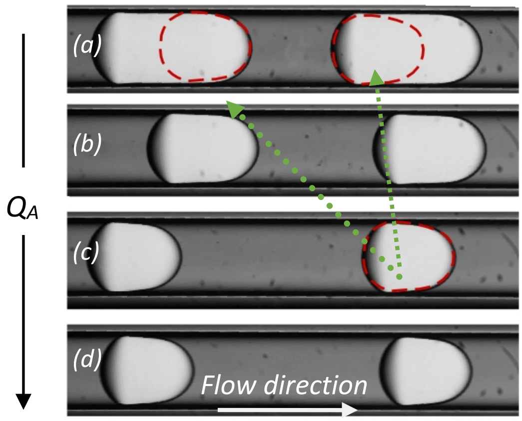


***Figure 4:*** *Effect of flow rate on the slug size for G2n and for* $Q_A$ (μL/min): *(a) 50, (b) 100, (c) 150, (d) 200.*

### *Effect of viscosity on the slug size*

The effect of viscosity on the slug size (length to width ratio, *L/D*) is presented in ***Figure 5*** for all the Newtonian fluids tested as a function of the aqueous to kerosene flow rate ratio ($Q_A/Q_K$). It is obvious that the slug size increases when the viscosity decreases. Also the slug size decreases with increasing flow rate. This trend is more pronounced in the relatively less viscous fluids.

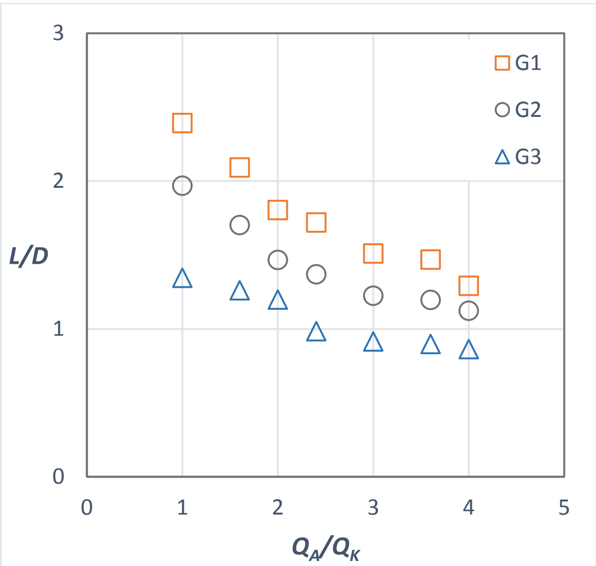


***Figure 5*** *Effect of the aqueous-phase viscosity on the slug size (Newtonian fluids).*

In ***Figure 6*** we compare the slug length (reduced with respect to the channel diameter) for a Newtonian fluid with the corresponding non-Newtonian one. Generally the slugs are sorter for the non-Newtonian case a fact that may be attributed to the higher viscosity of the liquid.

***Figure 6*** presents the slug *L/D* ratio as a function of the flow rate ratio $Q_A/Q_K$ of the two phases. The general trend is that the *L/D* ratio decreases with increasing viscosity. Moreover, the *L/D* ratio in all systems decreases with increasing aqueous solution flow rate, as it is also confirmed by Dessimoz et al. (2008) and Su et al. (2014).

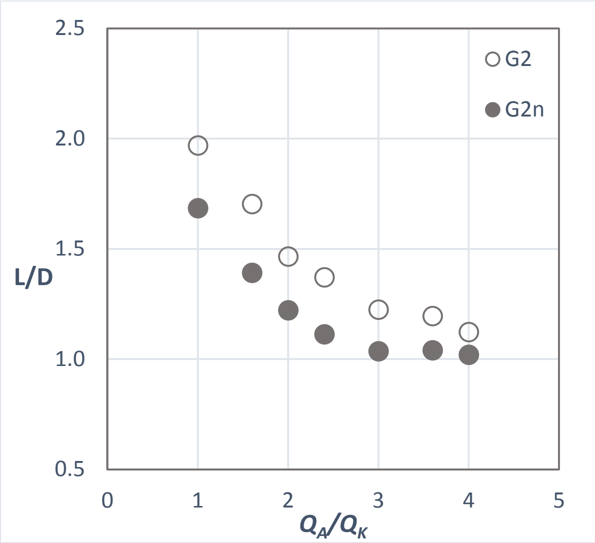


***Figure 6***: *Effect of the type of aqueous phase on the slug size.*

However, for higher flow rates (i.e. higher shear rates) where the viscosity of the non-Newtonian fluid attains its asymptotic value (***Figure 7***) the two liquids (*G2* and *G2n* or *G1* and *G1n*) result in slugs with the same size.

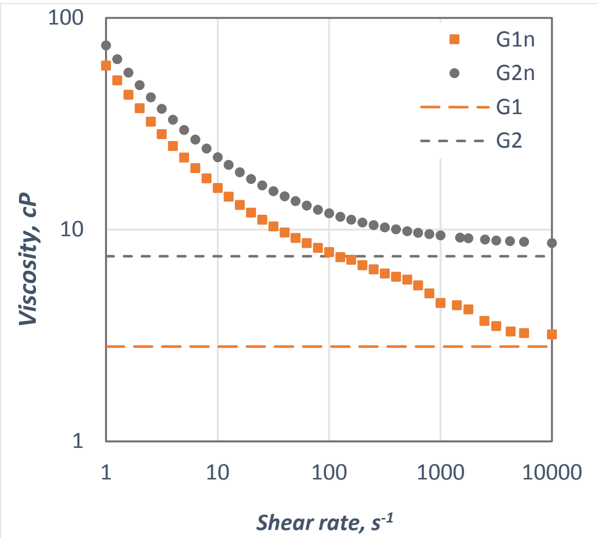


***Figure 7***: *Viscosity vs shear rate for the fluids used.*

Moreover, the *L/D* ratio of the kerosene slugs in both systems decreases with increasing aqueous-phase flow rate. An interesting observation is that for the *G2n* case, after a critical point (i.e. $Q_A/Q_K = 3$) the slug length remains practically constant (***Figure 6***). This can be also observed in ***Figure 4***. Thus for applications that have a given organic phase flow rate, a desired slug length can be achieved by choosing the appropriate aqueous phase flow rate.

The effect of the viscosity of the *non-Newtonian* continuous–phase on the size of the formed slugs is also examined. ***Figure 8*** shows that for the same flow rate the slug length decreases with increasing viscosity. As it has been already mentioned (***Figure 4***), the slug size of the *non-Newtonian* systems decrease with increasing aqueous-phase flow rate, as it can be depicted from ***Figure 8.***

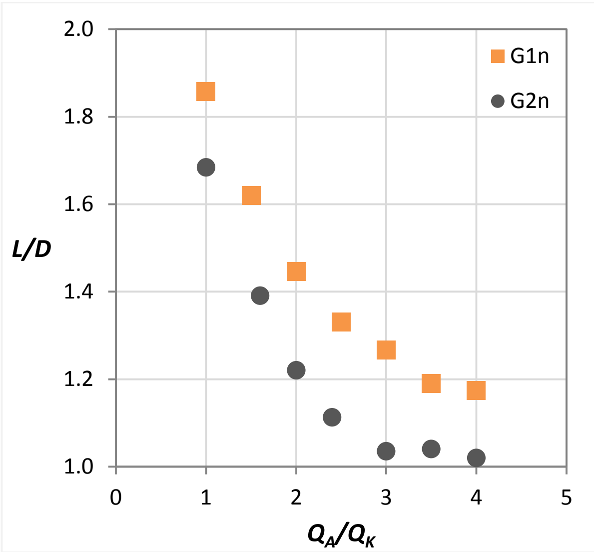


***Figure 8***: *Effect of the non-Newtonian aqueous-phase viscosity on the slug size.*

***Figure 9*** presents for the *non-Newtonian* fluids the effect of aqueous-phase viscosity on the size, the shape and the frequency of slugs. For a given flow rate of the aqueous-phase (i.e. $Q_A$ =100 μL/min), the less viscous liquid *G1n* forms relatively larger, more symmetrical and less frequent slugs than the *G2n*.

In ***Figure 10*** the effect of the aqueous phase viscosity on the frequency of the slugs is demonstrated. For the non-Newtonian *G2n* fluid and for the same flow rate the slug frequency increases and consequently the size of the slugs decreases compared with the *G1n*.

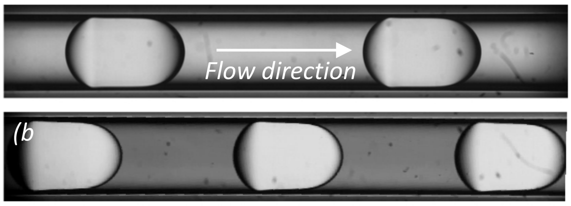


***Figure 9***: *Effect of aqueous-phase viscosity on the size, shape and frequency of slugs for (a) G1n, (b) G2n (non-Newtonian fluids, $Q_A$ =100 μL/min.*

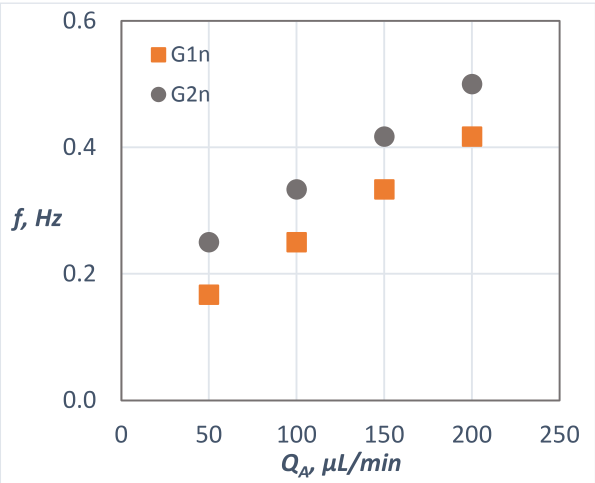


***Figure 10***: *Effect of the aqueous-phase viscosity on the slug frequency (non-Newtonian fluids).*

***Figure 11*** presents the frequency as a function of the aqueous flow rate of a Newtonian fluid and the corresponding non-Newtonian one (*G2* and *G2n*).

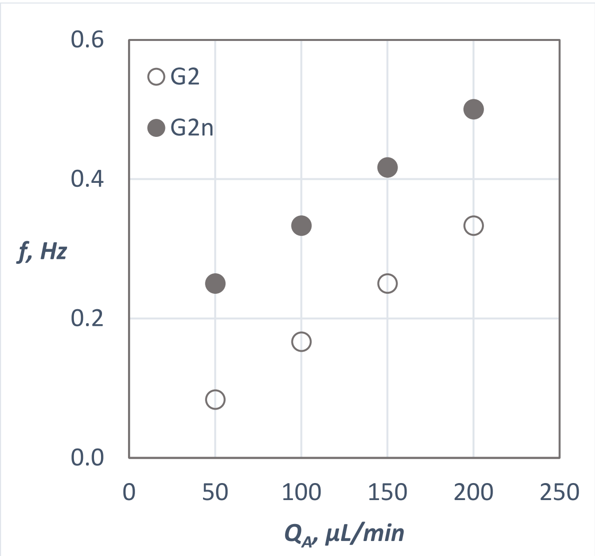


***Figure 11***: *Effect of the aqueous-phase viscosity (non-Newtonian and Newtonian) on the slug frequency.*

As it is expected, since the size of the slugs decreases with increasing viscosity, the higher viscosity results in higher slug frequency (a trend also observed in ***Figure 2***).

## 3. Conclusions

This study is the initial stage of a more extensive work that concerns the two-phase flow of an aqueous non-Newtonian and an organic liquid in microchannels. This type of flow is expected to be quite complicated since it is affected by the properties of the two fluids, the geometry and the material of the conduit as well as the type of the mixing section. Moreover it has not been adequately studied. In this stage to simplify the complicated problem we used a glass microchannel, a T-type mixing junction, only one flow rate of the organic Newtonian fluid (kerosene) and investigated the effect of the aqueous phase viscosity and flow rate on the shape, size and frequency of the formed slugs.

One flow pattern, namely slug flow, has been observed for all systems studied, irrespective of the fluid that initially fills the tube. In all cases, kerosene was the dispersed phase. From the experimental results, it can be extracted that:

- the non-Newtonian fluid results in smaller and more frequent slugs than the Newtonian one,
- the length of the kerosene slugs decreases by increasing either the flow rate or the viscosity of the non-Newtonian aqueous phase.

Consequently by rendering the aqueous phase non-Newtonian, i.e. by adding a small amount of xanthan gum, we can increase the interfacial area and enhance the mass transfer between the two phases. This observation might aid to the optimal design of two-phase equipment.

More work is needed and is currently in progress (i.e. experimental data for different geometrical characteristics of the microchannel, different organic phase etc.) in order to investigate the effect of all the design parameters on the characteristics of this kind of micro flow.

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