Experimental Investigation of Viscous Fingering Instabilities in Emulsions Displacements

Saraswathi Enjamoori., Jalel Azaiez* and Brij B Maini
Department of Chemical and Petroleum Engineering, The University of Calgary,
2500 University Drive NW, Calgary, Alberta, T2N 1N4 Canada
*E-mail: azaiez@ucalgary.ca

ABSTRACT
Experiments were conducted to analyze the viscous fingering instability that develops during the displacement of oil-in-water (O/W) emulsions. For this purpose, a horizontal rectilinear Hele-Shaw cell, which is an analogue for a homogeneous porous media, was used. Different emulsion solutions were prepared by dispersing different volume fractions of mineral oil in water using non-ionic surfactants. These emulsions were characterized systematically by examining their droplets size distribution and shear viscosity. Flow displacement experiments involved displacing the O/W emulsions by injecting water at different rates. The developments of the instability were characterized qualitatively by identifying the main mechanisms of finger development and growth. These results and the correlations between the properties of the fluids and the flow patterns were used to examine the flow dynamics and its effects on the viscous fingering instability. It was found that the development of the flow instability can be correlated with the rheological behaviour, which in turn depends on the emulsions' concentration and their internal microstructure.

1. INTRODUCTION

When a less-viscous fluid displaces another fluid of higher viscosity, instability can occur at the interface between the two fluids. The instability manifests itself in the form of finger-like patterns of the displacing fluid propagating through the displaced fluid, and is known as the viscous fingering instability. In horizontal miscible displacements, viscosity difference is the driving force whereas in vertical displacement processes, the instability may be also driven by a density difference. Such instability is encountered in a variety of natural and industrial applications and can have an important impact on the efficiency of those applications. In particular it is observed in a variety of oil recovery processes, solute transport in aquifers, chromatography and polymer processing.

A large number of studies dealing with the viscous fingering instability have been reported; most of these focused on displacements where both displacing and displaced fluids are Newtonian. The main controlling parameters in the Newtonian displacements would be the viscosity [1] or density ratio between the two fluids [2], injection rate [3, 4] interfacial tension between the two fluids [5] and the geometry of the cell [1]. The first studies to examine the viscous fingering instability date back to the 1950's with the original work of Hill [2] and of Saffman and Taylor[[6]. These were followed by numerous studies mainly in the 1980's that were discussed in the extensive review by Homsy [7]. Two reviews dealing with experimental perturbations by Maher et al.[8] and numerical modeling by Islam and Azaiez [9] have also appeared recently.

The past two decades witnessed an increased interest in analyzing the mechanisms of such instability in the case of non-Newtonian fluids such as polymer solutions and particle suspensions [10-13]. The fingering patterns observed in shear thinning polymer solutions were qualitatively different from those observed in Newtonian fluids [14]. Experiments carried out using shear thinning fluids showed that fingers grow mainly by shielding, spreading, tip splitting, dense-branching and skewering [15-18]. There are several studies on polymer solutions alone because of their complexity in their structure and rheological behavior. Experiments were carried out to understand the effects of polymer structures, molecular weights and solution concentrations on viscous fingering instability [17, 19, 20]. These studies allowed understanding, at least qualitatively, the effect of shear thinning on viscous fingering.

Later studies focussed on the explanation of observed finger patterns using theoretical and mathematical models [3, 21-23]. These theoretical investigations which were based on different types of rheological models, also revealed interesting morphological structures in the displacements of shear thinning fluids.

A limited number of studies focused on the importance of different perturbations sources associated with anisotropy due to the cell [24, 25], the presence of visco-elastic fluids [26] or fluids showing yield stress behavior [27, 28]. The more complex the rheological behaviour, the more complex the finger structures are. One of the less studied fluids exhibiting complex rheological behaviour, are emulsions which exhibit simple Newtonian to non-Newtonian behaviour. Interestingly, there is a real dearth of work on emulsion flows in spite of their importance in many applications. Such fluids are of particular importance in the oil industry where emulsions occur naturally as a result of the displacement of oil by water or are injected in processes of enhanced oil recovery. Even though emulsions share some important characteristics with non-Newtonian fluids such as the shear-thinning or visco-elastic behaviour, they in fact represent a class apart due to the complexity of their microstructure and compositions.

Interestingly, there is only one study reported in the literature that examined the viscous fingering instability in displacements involving emulsions [29]. In this study, the authors examined the flow in a radial Hele-Shaw cell for emulsions consisting of Silicone oil dispersed in aqueous Hydroxyl Propyl Methyl Cellulose (HPMC) solution. They observed crack-like finger to ramified finger patterns when the emulsion is displaced by an immiscible fluid. The authors related qualitatively the pattern transitions to the changes in the rheological properties of the emulsions and osmotic pressure difference between the injected fluid and the dispersion medium of the emulsion. However, they did not give any correlation between the imposed pressure and the injection rate or relate the physical behaviour of emulsions with the flow dynamics.

The present study aims at further improving our understanding of the fingering instability in emulsion displacements. It is proposed to examine the instability in the case of a rectilinear Hele-Shaw cell. The rectilinear geometry is different from the radial one adopted in the previous study [29] in that the latter involves a point source injection and contact interface that expands as the flow evolves while the former has a fixed initial interface defined by the cell width. Therefore, the development of the flow and the mechanisms of interactions as well as growth of the fingers can be fundamentally different. This is particularly true in the case of emulsions that exhibit complex rheological behaviour.

2. EXPERIMENTAL SETUP

2.1 Materials

Emulsions of oil in water (O/W) were prepared using a light mineral oil (trade name Marcol 7) supplied by Esso Imperial Oil, Canada Ltd. It is a highly refined colourless oil with very good chemical stability. The viscosity and the density of the oil at 23° C were 10.7 mPas and 862 kg/m3, respectively. The water used throughout the experiments was reverse osmosis de-ionized water. The surfactant used was Tween-65, a commercially available non-ionic surfactant also known as polyethylene glycol sorbitan tristearate. It is water soluble and has a hydrophilic-lipophilic balance (HLB) value of 10.5±1.0.

2.2 Sample Preparation Procedure

Two different concentrations of oil in water emulsions were prepared in batches of 200 ml each. The concentrations of oil were 40 and 60 vol% (surfactant free basis). The emulsion compositions are listed in Table 1.

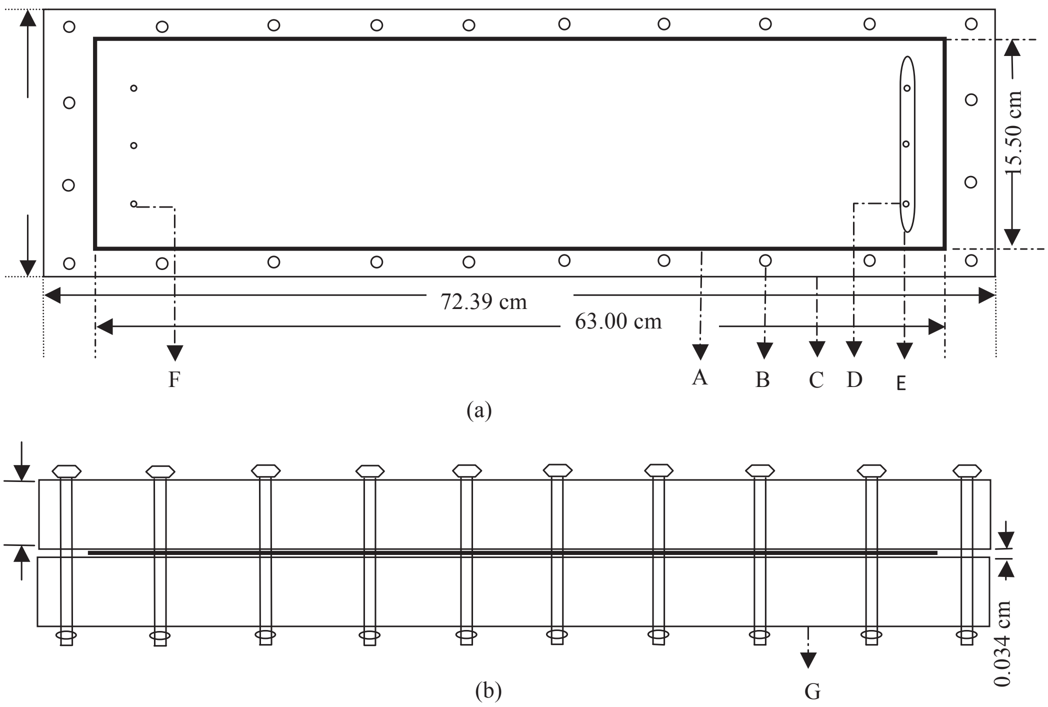
For the preparation of oil in water emulsions, the surfactant was dissolved in the aqueous phase by heating for about 15 to 20 min. Once the surfactant was completely dissolved, oil was added to the aqueous phase and heated for 7 to 8 min. The heating of the components reduced the oil viscosity and assisted in dispersing the oil droplets in the continuous water phase. The above mixture was then mixed in a blender for about 20 min at a fixed speed. The emulsions produced were quite stable with respect to coalescence. The emulsions were stored at 23±1° C.

Table 1. Details of emulsion samples

No	Concentration of oil in emulsion (Surfactant free basis) (V/V %)	Wt% of surfactant in continuous phase,
1	40%	8
2	60%	11

2.3 Hele-Shaw Cell

The flow displacement experiments were performed in a rectangular Hele-Shaw Cell [30]. The geometry of the cell is given in Figure 1.



Symbol	Name	Number	Comments
A	O-Ring	1	Buna-N
B	Bolts	24	Stainless steel
C	Top plate	1	Plexiglass
D	Injection ports	3	
E	Channel		
F	Production ports	3	
G	Bottom plate	1	Plexiglass

Figure 1. Schematic diagram of rectangular Hele-Shaw Cell (a) Top view, (b) Side view.

It consisted of two plane rectangular parallel glass plates made of plexiglass (72.39×20.32×5.08 cm3) that were separated by a narrow gap using a soft rubber o-ring (Buna- N, diameter 0.3175 cm) along the cell perimeter. The plates were fixed by 24 bolts and nuts.

On the top plate of the cell, three holes (diameter = 0.3175 cm) were drilled evenly along each short edge of the plate inside the o-ring. The centre hole at one end of the cell was used for the injection of the fluids during displacements while the centre hole at the other end was used for collecting the fluids flowing out of the cell. The use of single line injection/ production avoided the effect of different pressures in the three lines. The other two holes on both sides of the cell were used in filling the cell with the emulsion as well as in cleaning the cell between displacements.

The inner length, width and breadth of the channel were L=63 cm, W=15.5 cm and b=0.034 cm respectively. The cell was mounted on a light-table to assist in capturing images with better clarity. A differential pressure transmitter was connected across the inlet and outlet ends of the cell to record the pressure drop during the flow. The inlet of the cell was connected to a syringe pump that provides constant flow rates. The error involved in the flow rate was less than 4%.

A digital camera (Sony digital camera, Model No: DSC-P93A) was used to capture images of the cell during displacements. These captured images were analyzed using Image J software, which is a public domain Java image processing program that can run as an online applet or as a downloadable application, on any computer with a Java 1.4 or later virtual machine.

2.4 Displacement experiment procedure

Miscible displacement experiments were carried to examine the effect of changes in rheological behaviour of the displaced fluid (40% and 60% concentration emulsions), and injection rates (0.21 ml/min and 4.25 ml/min) on the viscous fingering instability.

For each experiment the following procedure was adopted. The displaced fluid (emulsion) was first injected into the Hele-Shaw cell with the syringe pump at very low injection flow rates to ensure that no gas bubbles were trapped inside the cell. The displacing fluid (water) was then injected into the cell at constant injection rate. For colour contrast between the displacing and displaced fluid, water was dyed with a red food colour.

The growth of the fingers was recorded periodically during the displacements using a camera. These captured images were analyzed using Image J software which is a public domain Java image processing program. The source code is developed and maintained by Wayne Rasband. It is provided through the National Institutes of Health (NIH) and is freely available.

3. CHARACTERIZATION OF EMULSIONS

The characterization of emulsions was carried based on their drop size distributions as well as their rheological properties. These measurements were conducted at 23±1°C after an elapsed time of approximately 5 hours from the initial time of emulsion preparation. In what follows, the results of drop size and rheological measurements for 40% and 60% concentration emulsions are presented.

3.1 Drop Size Measurement

The drop size analysis was carried out using a laser diffractometer Mastersizer 2000 (Malvern Instruments Co, UK) with Hydrosizer 2000S module. The results were obtained based on the intensity of the light scattered by the droplets in the range 0.02 to 2000µm. The size distribution calculations were based on the principle of Mie theory which predicts the way light is scattered by spherical particles and deals with the light diffraction pattern and using the standard software supplied with the instrument.

The measurements were performed by diluting 0.3g of emulsion in 8 ml of de-ionized water. The diluted sample was introduced into the laser beam using the Hydrosizer 2000s dispersion accessory. The measurements were carried out in triplicate and the averaged values are reported. The surface weighted mean diameter of 40% and 60% emulsion were 0.294µm and 0.159 µm, respectively. Note that the average drop size of 40% emulsion was larger than the 60% emulsion.

3.2 Rheological Measurements

A stress/rate controlled coaxial-cylindrical viscometer THERMO HAAKE RotoVisco-1 (Supplied byThermo Scientific) with a temperature controller, was used to measure the rheological properties of the emulsions. The start and end shear rate range was $\dot{\gamma} = 2 - 150 s^{-1}$.

Figure 2 shows the viscosity versus shear rate plot for 40% and 60% oil in water emulsions. The viscosity of 40% emulsion showed Newtonian behaviour with an average viscosity of 39m.Pas, whereas the 60% emulsion showed non-Newtonian shear thinning behaviour. The data is best fitted with a power-law model that is $\mu_{eff} = 1082.3\ \dot{\gamma}^{-0.43}$ with $R^2 = 0.99$. Where μ_{eff} is the effective viscosity and $\dot{\gamma}$ is the shear rate. These results show that with the increase in concentration of the dispersed phase, the rheological behaviour changed from simple Newtonian to shear thinning.

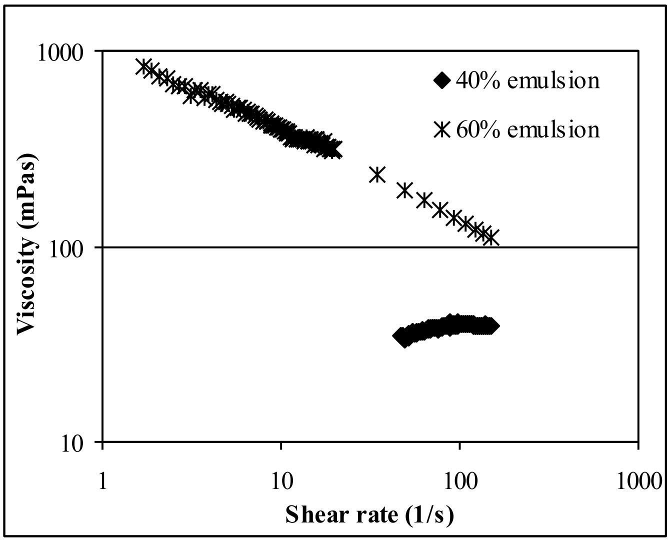


Figure 2. Viscosity versus shear rate plot for 40% and 60% concentration emulsions

4. RESULTS OF FLOW DISPLACEMENTS

In this section the displacement experimental results for both 40% and 60% emulsions are presented. In these displacements the effect of two different injection rates is shown. The fingering patterns observed are presented and discussed in terms of various fingering mechanisms involved. The results for the Newtonian 40% emulsion are discussed first followed by those for the non-Newtonian shear thinning 60% emulsion.

4.1 Qualitative Characterization

Experiments in which water displaces the emulsions were conducted at two different injection rates of 0.21ml/min and 4.25ml/min. Figure 3-6 show the sequential evolution of interface observed in emulsion displacements. The light space is the area filled with the emulsion and the red coloured region is the area occupied by the injected water. A sequence of four images at different times is presented.

4.1.1 40% emulsion displacements

Figure 3 and 4 show the results for 40% emulsion displacement at two different injection rates of 0.21ml/min and 4.25ml/min, respectively. At the lower injection rate of 0.21 ml/min, initially the interface started with three dense fingers. As the displacement progressed one finger dominated the flow suppressing the other two. This mechanism whereby one finger dominates its neighbours and out grows them is known as shielding [6]. Note that the fingers tend to have a diffuse structure which is particularly noticeable at later times.

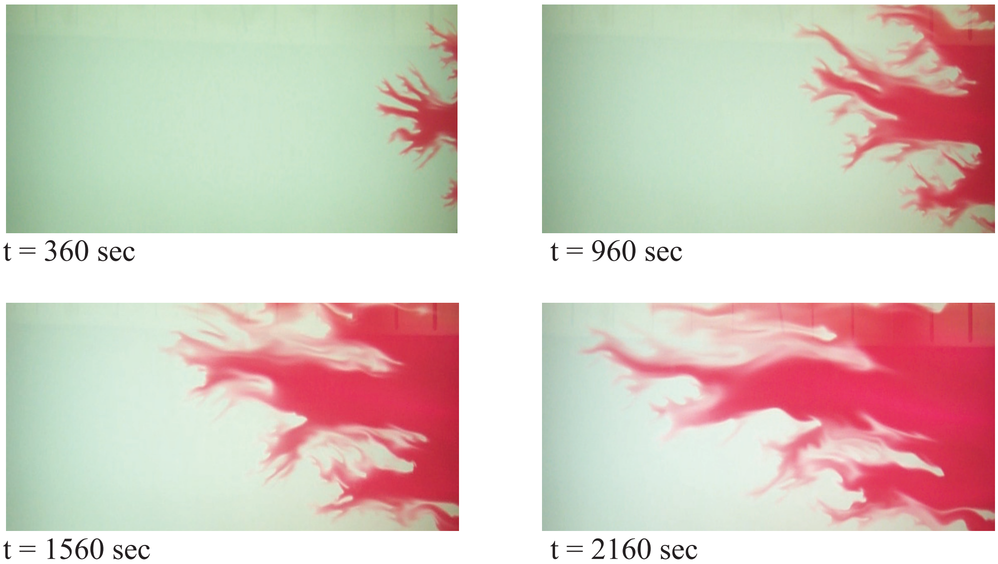


Figure 3. Displaceemnt of 40% emulsion at injection rate 0.21 ml/min

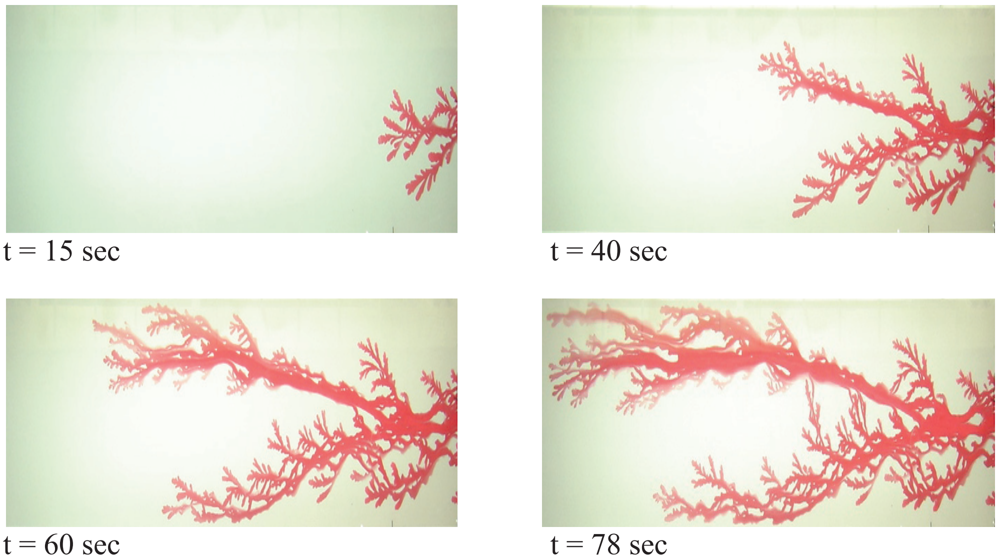


Figure 4. Displacement of 40% emulsion at injection rate 4.25 ml/min

The tips of the fingers were not smooth and became wider as the displacement progressed. This mechanism is known as spreading. The widely spread finger split into several thin fingers due to tip-splitting mechanism [31]. These fingers appear as faded side branch fingers. There is also noticeable diffusion of injected water into the displaced emulsion.

At a larger injection rate of 4.25 ml/min, the finger structures underwent considerable changes (Figure 4). The displacement started with two highly branched fingers out of which one became the dominant finger and moved ahead with continued branching. The dominant finger showed many side fingers known as side-branching [3]. Some of these branched fingers merged into the dominant finger and evolved through the main dominant finger as the flow progressed. This is known as coalescence [32]. The displacing fluid was diffusing into the displaced fluid in this test also but this effect was not as pronounced as in the case of the lower injection rate. As expected, the injected water reached the other side of the cell at a shorter time than in Figure 3. This will be examined later in the section dealing with the quantitative analysis.

4.1.2 60% emulsion displacements

Figures 5 and 6 show the results for 60% emulsion displacement at two different injection rates of 0.21 ml/min and 4.25 ml/min. The mechanisms responsible for the evolution of the fingers in these displacements are similar to those described earlier. In these displacements a single finger with many side branch fingers dominated the flow. However the dominant finger observed in this case is thinner compared to the 40% emulsion displacements. The interface between the two fluids is not smooth and showed side-branching mechanism. The growth of side branches increased with increase in the injection rate of the displacing fluid.

In these shear thinning fluid displacements the phenomena of shielding, tip splitting and coalescence were more pronounced (Figure 5). The fingers became more complex with many side branch fingers as the flow rate increased. Furthermore, when compared with the 40% emulsions, there is very little diffusion of the injected water into the emulsion.

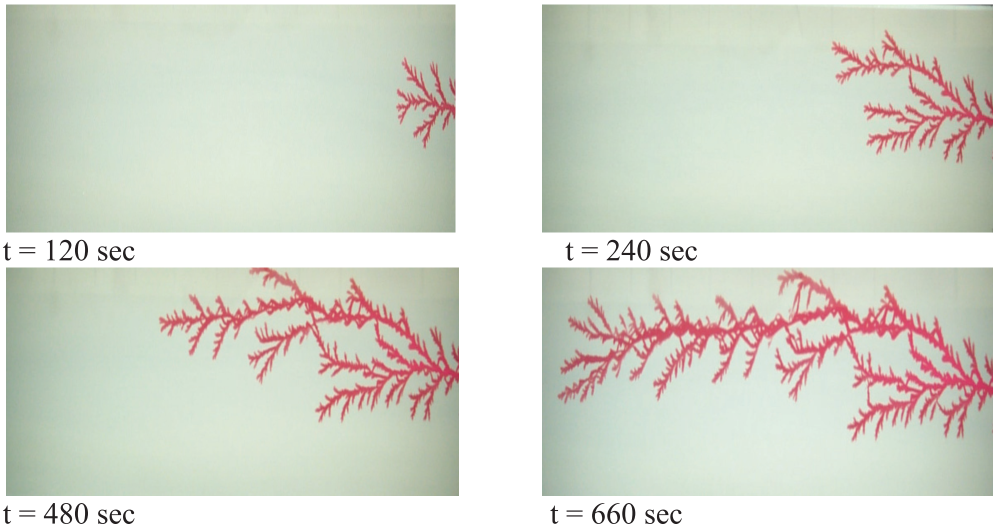


Figure 5. Displacement of 60% emulsion at injection rate 0.21 ml/min

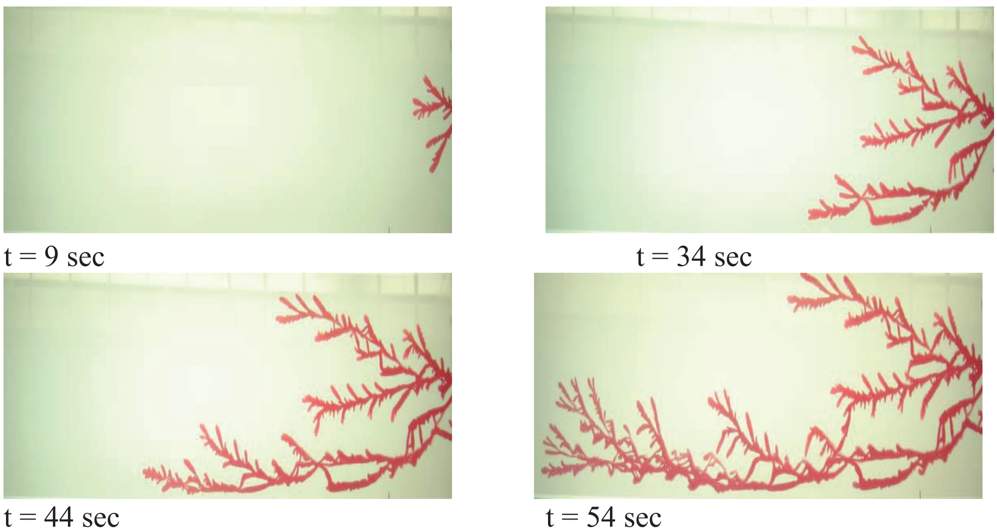


Figure 6. Displacement of 60% emulsion at injection rate 4.25 ml/min

4.2 Quantitative characterization

The growth of the finger structures and flow development will be further characterized through some quantitattive analysis. Figure 7 shows front length vs. the volume of fluid injected for 40% and 60% emulsion at different injection rates. As can be seen in the graph, the front length increases monotonically with the volume of fluid injected and also varies withh the injection rate. This shows that there is a significant effect of the injection rate on the front length, and therefore on the sweep efficiency of the displacement.

For the 40% emulsion, the front length increased with increasing injection rate while an opposite trend is observed in the case of the 60% emulsion. This difference may be explained by the different rheological behaviours of the two solutions. Indeed, the 40% emulsion shows a Newtonian behaviour whereby the viscosity does not change with the shear rate, or equivalently with the injection rate (see Fig. 2). Therefore, as the displacement velocity increases the viscous fingering instability is enhanced due to the diminished contribution of diffusive mixing and a stronger flow resistance. This leads to narrower and longer fingers that tend to move faster toward the production side.

The 60% emulsion on the other hand showed a non-Newtonian shear thinning behaviour, where the viscosity decreases with increasing shear rate (see Fig. 2). In this case, the effect of a larger injection rate that leads to a stronger flow resistance and weaker diffusion and hence faster growing fingers is still present. However, there is an additional countering effect associated with the decrease in the viscosity ratio between the two fluids as a result of the shear thinning behaviour of the displaced phase which tends to attenuate the instability and in turn the growth of the fingers. It appears that the destabilizing effects are more than compensated for by the decrease in the viscosity contrast, with the overall effect leading to a reduced instability and a decreased front length.

The displacements were also chacaterized by determining the breakthrough time, defined as the time where the injected water reaches the other side of the cell, and the sweep efficiency. The latter property is widely used in reservoir engineering to characterize how effective the displacement process is, and is defined as the fraction of the area contacted by the displacing fluid to the total area behind the displacement front.

In Table 2 the breakthrough time and sweep efficiency measurements are given. Here $E_{V,bt}$ is the volumetric sweep efficiency at breakthrough and $E_{V,avg}$ is the average of volumetric sweep efficiencies at eight evenly spaced times between the start of injection and the breakthrough.

The general equation used to determine the volumetric sweep efficiency is:

$$E_V = \frac{Q_i t}{60 L_f b w} \tag{1}$$

Where Q_i is injection rate (ml/min), L_f is finger front length (cm), t time from the start of displacement (sec), b gap of the channel (cm) and W width of the channel (cm). It should be noted that this equation is valid up to the breakthrough of the displacing fluid. In the case of $E_{V,bt}$, the time t in equation (1) corresponds to the breakthrough time *bt* representing the elapsed time between the start of the injection and the arrival of the displacing fluid to the production side of the cell.

The 40% emulsion showed the expected decrease in the sweep efficiency with the increase in injection rate. However, the shear-thinning emulsion displayed an increase in sweep with increasing injection rate. It appears that the higher shear rate caused by increased displacement velocity causes sufficient decrease in the apparent emulsion viscosity at the front to make the fingering less severe at higher injection rate.

Table 2. Quantitative analysis results for 40% and 60% emulsions.

Concentration of emulsion	Flow rate ml/min	t_{bt} sec	$E_{V,bt}$ %	$E_{V,avg}$ %
40%	0.21	2340	30	30
40%	4.25	76	20	20
60%	0.21	660	8	10
60%	4.25	54	14	18

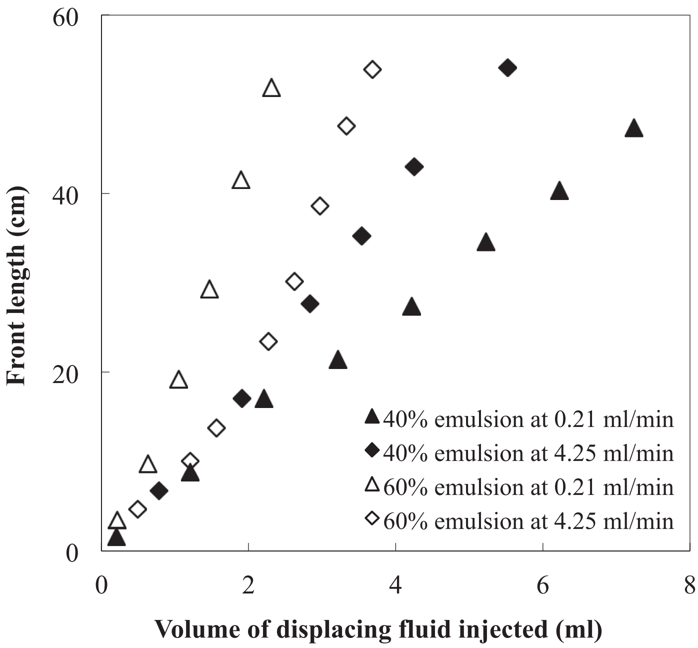


Figure 7. Front length (y) versus volume of fluid injected (x) for 40% and 60% emulsions at two different injection rates.

5. CONCLUSION

The viscous fingering instability in the displacement of oil in water emulsions was investigated. The choice of two different concentrations i.e., 40% and 60% (v/v %) emulsions allowed isolating the effects of the Newtonian and non-Newtonian shear thinning fluid behaviour on the instability. Experiments were carried at two different injection rates of 0.21 ml/min and 4.25 ml/min.

In the experimental study, water was used as the displacing fluid and emulsions as the displaced fluids. These emulsions were characterized in terms of their drop size and rheological measurements. The average drop size of 40% emulsions was larger than 60% emulsion and their rheological measurements revealed that the 40% emulsion was behaving as a Newtonian fluid while the 60% emulsion showed a non-Newtonian shear thinning behaviour.

The displacement experiments showed that as the rheological behaviour of the displaced fluid changed, there is a significant change in their finger structures. The width of the dominant finger decreased and the amount of branching increased. For both emulsions, an increase of the injection rate led to more complex finger structures.

The effect of inreasing the injection rate on sweep efficiency was different in the two emulsions. The Newtonian emulsions showed decreased sweep at higher injection rate. This is the expected behavior since the fingering is expected to increase with increasing displacement velocity. This was not the case in diplacements of the non-Newtonian emulsion. Here the sweep efficiency improved with increasing velocity.

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