EXPERIMENTAL INVESTIGATION ON LIQUID-LIQUID DISPLACEMENT OF VISCOELASTIC MATERIALS IN CAPILLARY TUBES

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Abstract. *The displacement of a fluid by liquid injections occurs in some practical applications like oil recovery in porous media and cementation of drilling wells. For a good understanding of these problems, it is extremely important to analyze the effect of the capillary number, Reynolds number and viscosity ratio in these flows. Furthermore, the presence of non-Newtonian liquids in these processes is quite common. Therefore, the study of rheological properties, like viscoelasticity, is in fact desirable. The fractional mass deposited on the tube wall and the shape of the interface on liquid-liquid displacement has been studied previously, experimental and numerically, by Soares, Souza Mendes and Carvalho (2005). However, the liquids were Newtonian and only the effects of capillarity and viscosity ratio were investigated. Hence, the goal of the present work is an experimental analysis of viscoelastic effects on the fractional coverage for the case where the Newtonian liquid is displaced by the viscoelastic liquid and for the case where the viscoelastic liquid is displaced by the Newtonian liquid.*

Keywords*: Liquid-liquid displacement, viscoelastic liquids, experimental investigation, fractional coverage*

1. Introduction

The present work analyzes experimentally the displacement of a fluid in the interior of a capillary tube by the injection of another fluid, which is immiscible with the first. The tests were performed using a viscoelastic liquid as either a displacing or displaced material. Practical applications include the cementation process of production and injection wells and the flow through porous media during enhanced oil recovery. An overview of selected oil recovery processes shows that hydrolyzed polyacrilamide and biopolymers, as xanthan gun, are commonly pumped into oil reservoir in order to aid oil recovery by providing mobility control to the injection water, as extensively discussed in the articles of Dreher and Gogarty (1979), Johansen (1979) and Khagram, Gupta and Sridhar (1985). These materials are non-Newtonians, presenting high viscoelastic effect. Hence, the analysis of these properties is clearly of interest for improvements of oil recovery operations.

It is important to understand the mechanism of liquid displacement and determine the amount of liquid that is left behind adjacent to the wall. The configuration of the interface between the two liquids depends on the force balance near the interface, which is, in this case, determined by the Reynolds number, Capillary number, Viscosity ratio and rheological properties.

Most of the related work found in the literature deals with the case of a gas displacing a viscous liquid, going back to the pioneer work of Fairbrother and Stubbs (1935) and Taylor (1961). In the experiments reported in these early papers, the Reynolds number was kept small enough to assure negligible inertial effects. The main goal was to determine the fraction of mass deposited on the tube wall *m*, which, with the aid of the mass conservation principle, can be written as a function of the velocity of the tip of the interface *U* and the mean velocity $\overline{u}$ of the liquid ahead of the gas-liquid interface, as show Eq. (1).

$$m = \frac{U - \overline{u}}{U} \quad (1)$$

Taylor (1961) studied the dependence of the mass fraction on the *capillary number* $Ca = \mu U / \sigma$, where μ and σ are the liquid viscosity and surface tension, respectively. His analysis indicated that the amount of liquid deposited on the wall rises with the interface speed, and that *m* tends asymptotically to a value of 0.56 as *Ca* approaches 2. Working on the same problem, Cox (1962) studied the mass fraction over a wider range of the capillary number, and also observed that *m* reaches an asymptotic value at high capillary number. However, he showed that this asymptotic value was 0.60 as *Ca* approached 10. Using the lubrication approximation, Bretherton (1961) derived a theoretical correlation between the mass fraction and the Capillary number, and the agreement between his predictions and Cox's experiments is good in the range of $10^{-3} \le Ca \le 10^{-2}$.

The effect of shear thinning behavior of the displaced liquid by gas injection was studied by Poslinski and Coyle (1994). They used the Finite Element Method to solve the two-dimensional model of the flow. Kamisli and Ryan (1999) performed experiments and showed that the thickness of the deposited liquid layer falls with the power-law index. They presented a singular perturbation analysis to model this situation, but their predictions followed the opposite trend of the experimental results.

The penetration of a long gas bubble in a viscoelastic liquid was first studied experimentally by Huzyak and Koelling (1997). They were interested in identifying the influence of viscoelastic behavior on the fraction of mass deposited on the tube wall. The experiments were performed with a highly elastic liquid with a constant shear viscosity. The results were presented in terms of capillary number *Ca* and Deborah number *De*. They found that the fractional mass deposited on the wall begins to increase, relatively to Newtonian fluid, for $De \ge 1$ and continues increasing over the entire range of *De* analyzed. As will be presented later by the present work, this result is in qualitative agreement when a Newtonian liquid is used to displace another viscoelastic fluid. Following the work of Huzyak and Koelling (1997), Gauri and Koelling (1999) analyzed the kinematics of the flow near the free surface using Particle Tracking Velocimetry (PTV).

Articles dealing with the analysis of liquid-liquid displacement in capillary tubes are much scarcer. One of these few papers is given by Goldsmith and Mason (1963) who report experimental results on the amount of displaced liquid left on the tube wall as a function of different parameters when two Newtonian liquids are used. In their experiments, the displacing material is a long drop of a viscous liquid. The results showed that the mass fraction rises as the viscosity ratio $N_\mu = \mu_2 / \mu_1$ is decreased, where the index 1 refers to the displacing fluid and the index 2, to the displaced fluid. This trend agrees with the theoretical predictions and experimental data presented in a preview work by Soares, Souza Mendes and Carvalho (2005).

Continuing the work of Soares, Souza Mendes and Carvalho (2005), in the present work, the steady displacement of a viscous liquid by a long drop of another viscous liquid in a capillary tube is analyzed experimentally to verify the influence of the viscoelastic behavior in this kind of flow. A thin layer of the displaced liquid is left behind on the tube walls, as illustrated in Fig. 1. In the Figure, *Ro* is the radius of the tube and R_b the radius of the cylindrical portion of the interface. The experiments consisted of visualization of the tip of the interface to examine its shape and the amount of displaced liquid left on the tube wall.

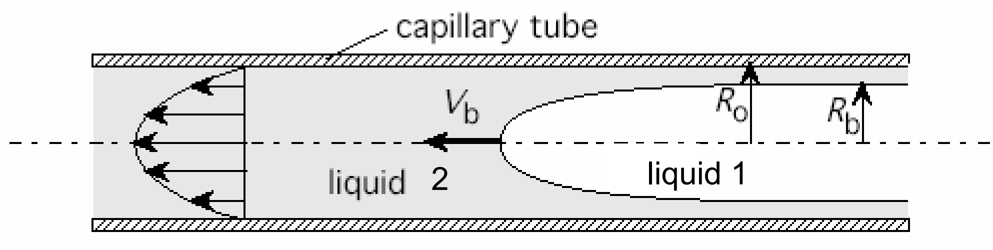

Figure 1. Schematics of the liquid-liquid displacement in the capillary tube

2. Experimental apparatus

The experimental setup used in the visualization experiments is sketched in Fig.2. Initially, the displacing liquid (Liquid 1) is stored in reservoir (A), and the displaced liquid (Liquid 2), in reservoir (E). Collecting Tanks (B) and (C) are auxiliary tanks used in the process of eliminating air bubbles in the system, as explained below.

The flow was controlled by cutoff valves (V1) to (V5) and by the gate valve (R1). The glass capillary tube was *L*=1.5 m long, and the inner and outer diameters were *Do*=5 mm and D_{ext}=7 mm, respectively. The tube was mounted inside a plexiglas box (D) filled with glycerin in order to minimize distortions of the interface image. A charge-coupled device (CCD) camera mounted at a right angle to the side wall of the plexiglas box was used to get the image of the interface as it traveled through the glass capillary. The camera was connected to a VCR to record the images.

The selection of the liquids was crucial to the experiments. The pair of liquids used had to be immiscible and with refractive indexes sufficiently different from each other in order to render the interface visible. Furthermore, the densities had to be as close as possible to minimize buoyancy effects. The buoyancy force is proportional to the magnitude of the density difference $\Delta\rho = \rho_1 - \rho_2$, and to the square of the drop diameter, D_b. As can be seen in the articles of Cox (1962) and Bretherton (1961), for gas-liquid displacement, the importance of buoyancy relative to viscous forces is small when:

$$\frac{(\Delta\rho)gD_b^2}{\mu U} << 1 \qquad (2)$$

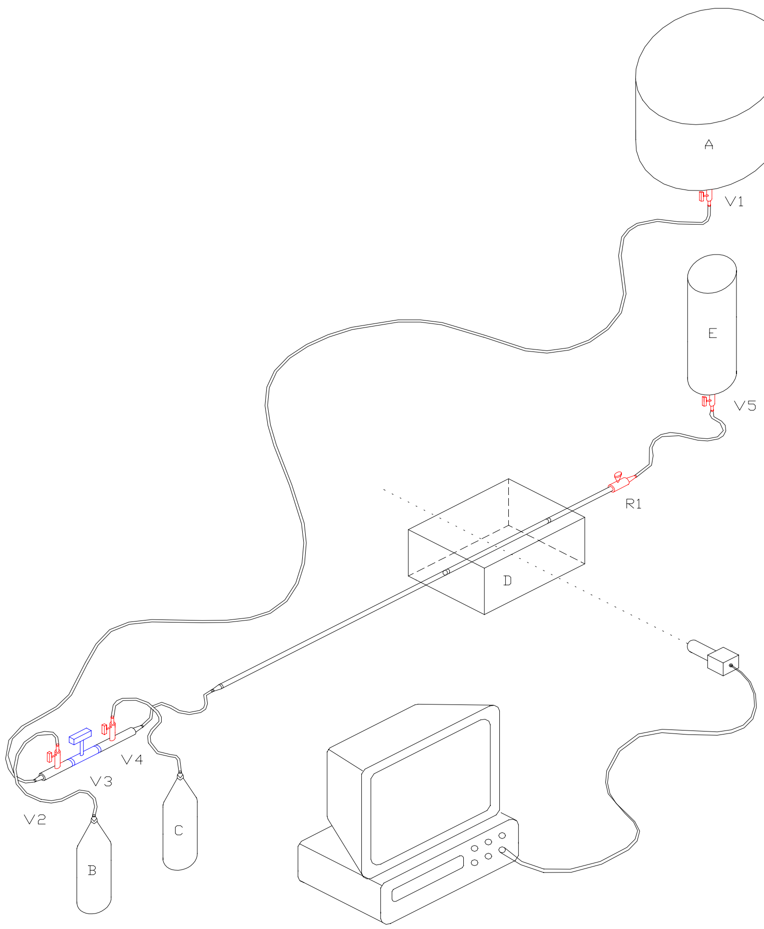

Figure 2. Schematics of the apparatus

where g is the acceleration due to gravity. This ratio was useful in providing design guidelines for the experiments, although the characteristic viscous force employed is less than satisfactory for liquid-liquid displacement. Two different tipe of Newtonian liquid was used. The first, used as either the displacing or the displaced material, was a soybean oil. Its density was $\rho = 915$ kg/m^3 and its viscosity $\mu = 50x10^{-3}$ Pa.s. The second was a mixture of water and ethanol, with $\rho = 915$ kg/m^3 and $\mu = 2,5x10^{-3}$ Pa.s. The viscoelastic liquid was a solution of PEG (polyethylene glycol, MW 56000 g/mol) and PEO (oxide of polyethylene glycol, MW 4000000 g/mol) in water. The viscosity level could be selected by changing the PEG concentration, while the elastic intensity could be controlled by changing the PEO concentration. The density of the PEG and PEO aqueous solution was virtually constant over the range of concentration explored, namely, $\rho = 1030$ kg/m^3.

Before starting each visualization experiment, the glass capillary has to be filled with the displaced liquid (Liquid 2), stored in Reservoir (E). This is done by opening valves V4, V5, and R1, while keeping valve V3 closed. This causes the liquid to flow from Reservoir (E) to the collecting tank (C). This flow is maintained until all the air bubbles are removed from the capillary tube. A similar procedure is followed to fill the line from reservoir (A) to valve V3 with the displacing liquid (Liquid 1). Again, the flow is maintained until all the air bubbles are removed. To start the experiment, reservoir (E) is disconnected from the capillary tube and valve V3 is opened, with V2 and V4 closed. The tip of the interface starts traveling through the capillary tube and its velocity is controlled by the gate-valve R1. The CCD camera captures the image of the interface as it passes through the visualization box. The amount of Liquid 2 that remains on the tube wall is determined a posteriori by an image analysis. The level variation at Reservoir (A) during each experiment is negligible because the transversal area of the reservoir is sufficiently large, rendering the pressure gradient essentially constant during each experiment.

Because the liquids had different viscosities, the pressure loss along the capillary changes as Liquid 1 takes over the tube. This change in pressure loss would lead to a change in the drop speed as it travels through the capillary. This is avoided by assuring that most of the overall pressure loss occurs in the gate valve, so that the pressure loss along the capillary is always negligible. Therefore, any variation that occurs in the later causes no appreciable change in the overall pressure loss of the system, and thus the interface always travels with constant speed. The interface velocity *U* is measured by recording the time that it takes to travel between two axial positions marked on the tube. The maximum interface velocity that could be measured with accuracy was of about 0.15 m/s, which determined the maximum capillary number that could be obtained in the experiments. The surface tension for each pair of liquids is measured by carefully placing a layer of the lighter liquid at the top of the surface of the heavier liquid previously placed in a beaker, and then using a Lauda ring tensiometer to determine the interfacial tension. After each run, the tube is rigorously cleaned to remove all contaminants. This procedure is needed to avoid changes in surface tension.

2.1. Fluids characterization

Two samples of viscoelastic solutions were used in the experiments. The first was composed by 20%, in weight, of PEG and 0,1% of PEO and the second was prepared with 30% of PEG and 0,21% of PEO. Since the PEO has a rather small solubility in water, the samples were mixed in a rotating cylinder reservoir with a really slow revolution for about three days, to avoid air bubbles and aggregates.

Rheological tests were performed in order to characterize the non-Newtonian liquids. The dependence of the shear viscosity, η_S, and the first normal stress difference coefficient, Ψ_1, with the shear rate was obtained with a rotational rheometer. The dependence of extensional viscosity, η_E, with extensional deformation rate was performed with a extensional rheometer.

Figure 3 shows data of shear viscosity with respect to shear rate. It is clear a shear-thinning behavior for values of shear rate smaller than 50 s^{-1}. However, the shear viscosity tends asymptotically to a value of 0.075Pa.s when the shear rate reaches 100 s^{-1}. The sample composed by 20% of PEG and 0.1% of PEO showed a constant shear viscosity of 0.0275 Pa.s over the entire range of shear rate tested.

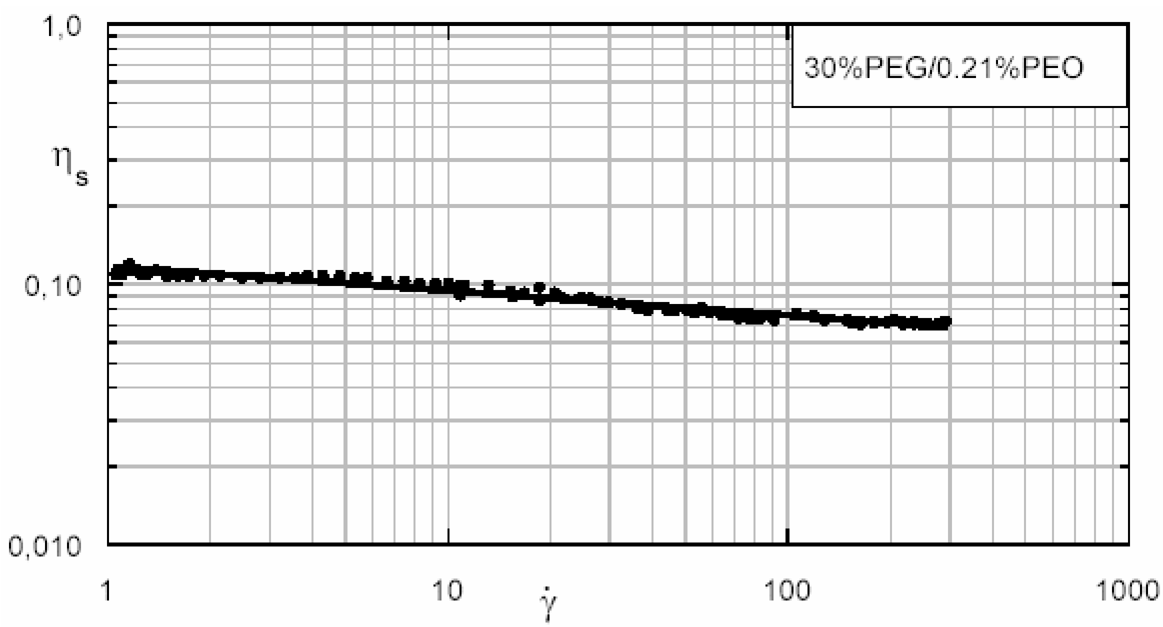

Figure 3. Shear viscosity dependence with shear rate for a solution of 30% of PEG and 0,21% of PEO in water.

Figure 4 shows data for the first normal stress difference coefficient, Ψ_1, with respect to shear rate for the sample composed with 30% of PEG and 0.21% of PEO. It could be observed that Ψ_1 decreases significantly in the range of $10 \leq \dot{\gamma} \leq 300$. The first normal stress difference coefficient for the sample composed with 20% of PEG and 0.1% of PEO is rather smaller than the values presented in the Fig. 4 and the equipment used was not capable to perform measurements for this sample.

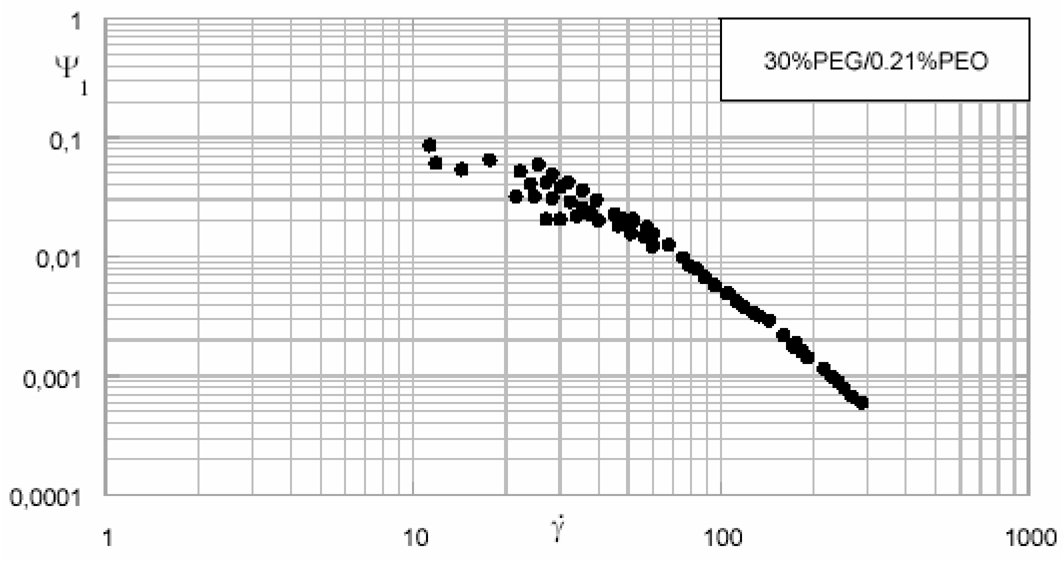

Figure 4. First normal stress difference dependence with shear rate for a solution of 30% of PEG and 0,21% of PEO in water

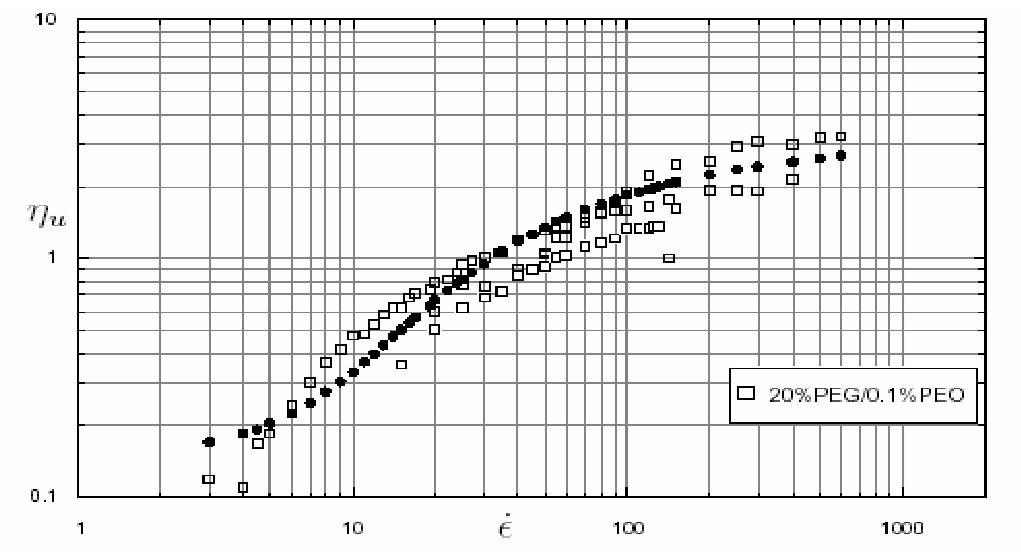

Figure 5. Extensional viscosity dependence with extensional rate for a solution of 20% of PEG and 0,1% of PEO in water

The extensional viscosity dependence with extensional rate for the viscoelastic samples are presented on Fig. 5 and Fig. 6. It is observed the extensional thickening effect for the two solutions tested. Furthermore, the extensional viscosity analysis suggests a Newtonian level when the extensional rate tends to quite small and very large values.

After an overview on rheological tests for the viscoelastic samples and observing the high shear thinning behavior for the first normal stress difference coefficient, it can be expected that the extensional viscosity, η_E, must become the principal non-Newtonian parameter as the deformation rate is incremented. Rheological properties dominated by extensional viscosity is quite common for polymeric solutions used in oil recovery processes, Khagran, Gupta and Sridhar (1985).

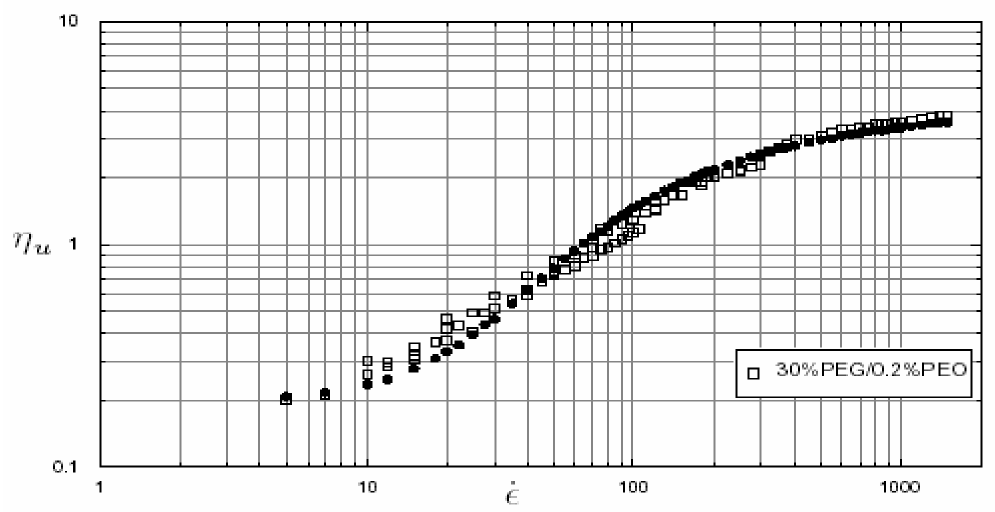

Figure 6. Extensional viscosity dependence with extensional rate for a solution of 30% of PEG and 0,21% of PEO in water

3. Results

The amount of Liquid 2 that remains on the capillary wall is usually reported in terms of the mass fraction of liquid that is not displaced *m*, or simply by the liquid film thickness left on the wall h_∞, as shown on Fig. 7. The two forms are related by

$$m = \frac{\text{mass left on the wall}}{\text{total mass}} = 1 - \frac{\text{displaced mass}}{\text{total mass}} = 1 - \left(\frac{D_b}{D_0}\right)^2 = 1 - \left(1 - \frac{2h_\infty}{D_0}\right)^2 \tag{3}$$

where $h_\infty = (D_0 - D_b)/2$ is the layer thickness of Liquid 2 left on the wall.

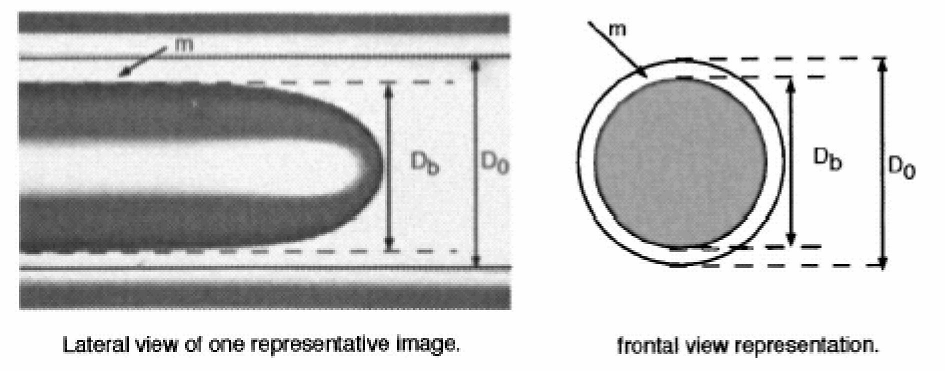

Figure 7. Representative image of the interface

The experimental data were obtained for Reynolds numbers small enough to assure negligible inertial effects. The main goal was to study the effect of viscoelastic properties on the mass fraction, *m*, and on the shape of the tip of the interface. The viscoelasticity reported by the Deborah number which is a relation between a characteristic time of the liquid and a characteristic time of the flow can be related by

$$D_e = \frac{\Psi_{10}}{2\eta_0}\left(\frac{U}{D}\right) \tag{4}$$

where $\Psi_{10} / 2\eta_0$ is the characteristic time of the liquid related to rheological properties measured in small shear rates and U / D is a characteristic shear rate of the flow.

In order to validate the experimental procedure, the results of Taylor (1961) for gas-liquid displacement were employed. Figure 8 shows Taylor's experimental data together with the present experimental results of liquid-liquid displacement for two Newtonian liquids with $N_\eta = 20$. The agreement is quite good over the range of capillary numbers explored. The viscosity ratio $N_\eta = 20$ was shown to be large enough to reproduce the limiting case of gas-liquid displacement.

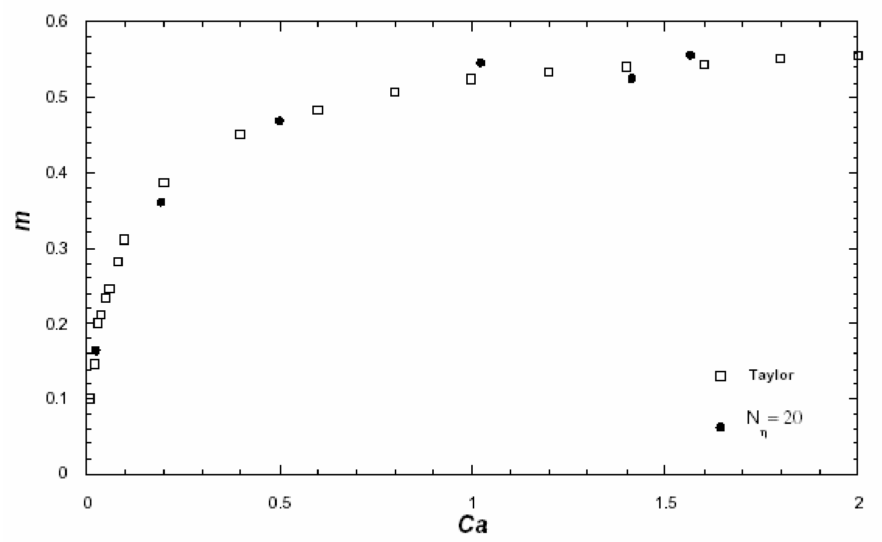

Figure 8. Fraction of mass deposited on the tube wall as a function of capillary number. The black dots are the results obtained by the present work for two Newtonian liquids and the squares dots are Taylor´s data for the gas-liquid displacement.

The image of the interface for the solution composed by 20% of PEG and 0.1% of PEO displacing the Newtonian liquid is compared with the image of the interface when two Newtonian liquids are used for a fixed capillary number and viscosity ratio. The images suggest that the interface tends to be flatter when a viscoelastic solution is used as the displacing liquid.

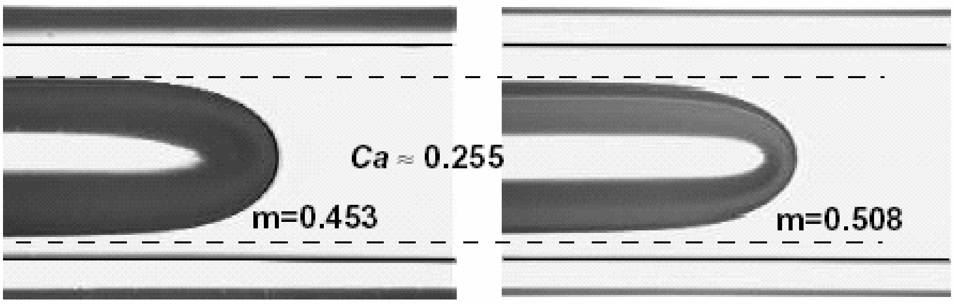

Figure 9. Images of the tip of the interface for a fixed capillary number, *Ca=0.255*, and viscosity ratio, $N_\eta=1.8$. The left image is for the solution of 20% of PEG and 0.1% of PEO displacing a Newtonian liquid and the right image is for the case when two Newtonian liquid are used.

Figure 10 shows the fraction of Liquid 2 left on the tube in function of capillary number for viscosity ratio fixed in *1.8*. The black dots are data for the case when the solution of 20% of PEG and 0.1% of PEO is used as the displacing liquid and the white dots are results when two Newtonian liquids are used. The results suggest that the fraction of mass, *m*, reaches *0.43* asymptotically as the capillary number approaches *0.2,* when the viscoelastic solution is the displacing liquid. While for the case when two Newtonian liquids are used, the fraction of mass continues to increase over the entire range of capillary number analyzed. Hence, the experiments indicate that a viscoelastic solution is more efficient in oil recovery in capillary tubes.

The results presented on Fig. 11, Fig. 12 and Fig. 13 are related to the case when the solution composed by 30% of PEG and 0.21% of PEO is displaced by the Newtonian liquid. Respectively, Fig. 11 and Fig. 12 present the images of the tip of the interface and their shapes, comparing the viscoelastic interface with that observed when two Newtonian liquids are used at fixed capillary number, $Ca = 0.26$, and viscosity ratio, $N_\eta = 1.8$. The images show a quite sharper interface for the case of a viscoelastic displaced liquid.

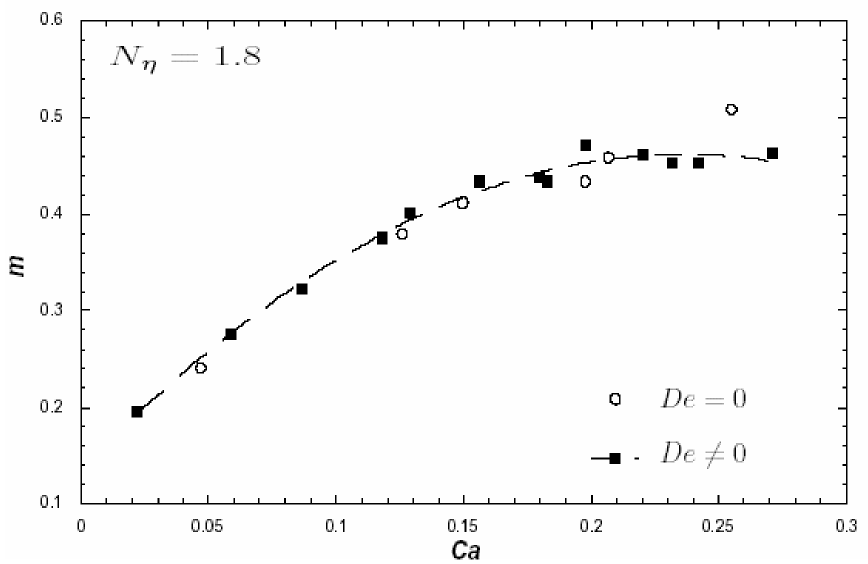

Figure 10. Fraction of mass deposited on the tube wall as a function of capillary number for a fixed viscosity ratio. The white dots are results for the displacement of a Newtonian liquids by another Newtonian liquid and the black dots are data for the case when the solution composed by 20% of PEG and 0.1% of PEO is used to displace a Newtonian liquid.

Finally, the fraction of mass deposited on the tube wall with respect to capillary number is presented on Fig. 13 for fixed viscosity ratio, $N_\eta = 1.8$. The black dots are data for the case when the viscoelastic solution is used as displaced liquid and the white dots are data for two Newtonian liquids flowing within the capillary tube. Over the entire range of capillary number analyzed, the results indicate a thicker liquid film when the viscoelastic solution is displaced by a Newtonian liquid. This is in agreement with the experiments reported from Huzyak and Koelling (1997) for gas-displacement of viscoelastic liquids in capillary tubes. They found that the fraction of mass increase with respect to Newtonian for Deborah number larger than unit. Hence, as in gas-liquid displacement, the process of oil recovery in capillary tubes is less efficient when the displaced liquid present viscoelastic behavior.

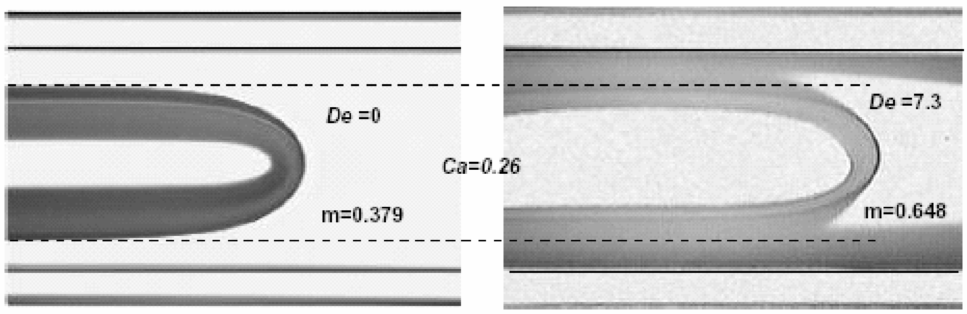

Figure 11. Images of the tip of the interface for a fixed capillary number, *Ca=0.26*, and viscosity ratio, $N_\eta=1.8$. The right image is for the solution of 30% of PEG and 0.21% of PEO displaced by a Newtonian liquid and the left image is for the case when two Newtonian liquid are used.

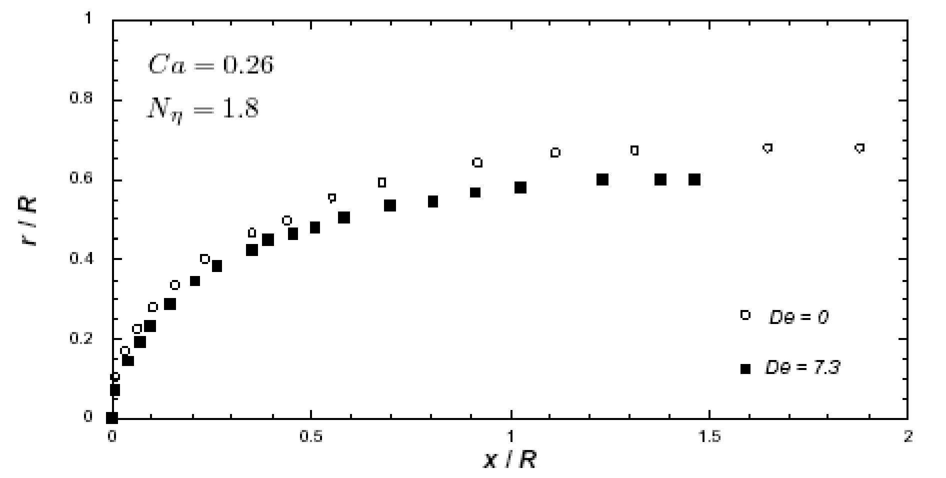

Figure 12. Shape of the tip of the interface. The black dots delimit the tip of the interface when the solution of 30% of PEG and 0.21% of PEO is displaced by a Newtonian liquid and the white dots delimit the tip of the interface when the displacement occurs with two Newtonian liquids.

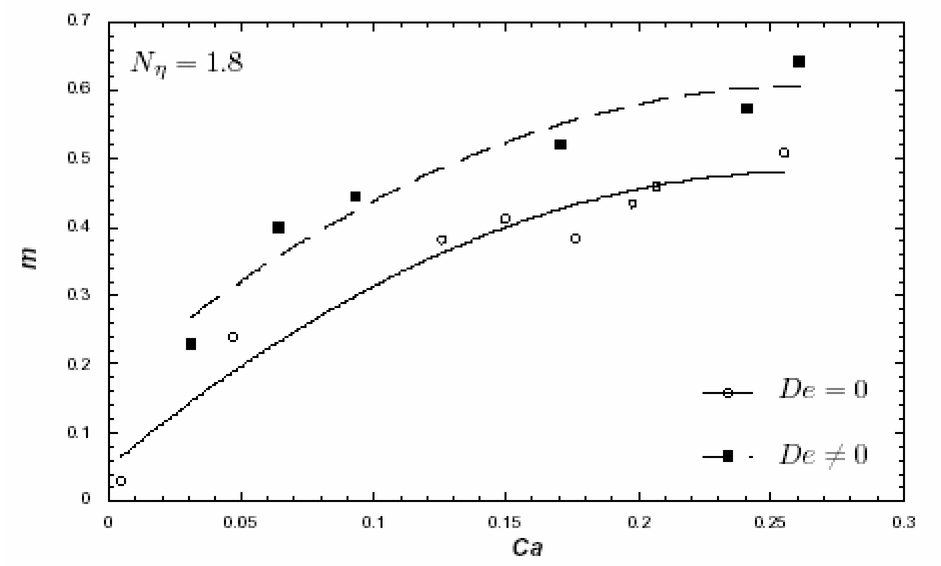

Figure 13. Fraction of mass deposited on the tube wall as a function of capillary number for a fixed viscosity ratio. The white dots are results for the displacement of a Newtonian liquids by another Newtonian liquid and the black dots are data for the case when the solution composed by 30% of PEG and 0.21% of PEO is displaced by a Newtonian liquid.

4. Conclusions

Experiments of liquid-liquid displacement in capillary tube using viscoelastic solutions as either the displacing or the displaced material was presented. The experimental apparatus was built in order to capture the image of the tip of the interface for analyzing its shape and the fraction of the displaced liquid mass attached on the wall.

Recent articles are found in the literature which analyze liquid displacement in tubes. However, these are limited to gas-liquid displacement or to liquid-liquid displacement of Newtonian materials. Thus, the main contribution of the present work was the study the liquid-liquid displacement in tubes for non-Newtonian solutions presenting high extensional viscosity. These solutions are commonly pumped into oil reservoir in order to aid oil recovery. Nevertheless, the range of capillary number analyzed was rather small and a more accurate study is still necessary.

The validation of the experimental apparatus was performed confronting the results for two Newtonian liquids with large value of viscosity ratio with data from Taylor (1961), obtaining a quite good agreement.

Visualizations were performed to confront the shapes of the tip of the interface for two Newtonian liquids and for the case when viscoelastic solutions was used as either displacing or displaced liquids at fixed capillary number and viscosity ratio. The images indicate that the interface became flatter when the viscoelastic solution is used as displacing liquid, while, it became sharper when the viscoelastic solution is displaced by a Newtonian liquid.

When the viscoelastic solution is used as the displacing liquid, the images analysis suggests that the fractional mass, *m*, tends asymptotically to *0.43* as the capillary number approaches *0.2,* while, it continue to increase for the case of two Newtonian liquids witch retaining a thicker liquid film attached on the wall. Finally, in respect to Newtonian liquid-liquid displacement, the visualizations indicate that the displacement of viscoelastic solutions by Newtonian liquids implies an increasing on the fraction of mass left behind, over the entire range of capillary number analyzed.

5. Acknowledgements

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