Oil displacement in capillary tubes using viscoelastic fluids

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Abstract: *Capillary invasion of liquid is a spontaneous phenomenon that occurs at length scales smaller than the capillary length. By means of capillary action, numerous applications have been developed. We investigate the time-dependent displacement of a Newtonian oil using viscoelastic polyethylene oxide solutions. The setup implies a capillary tube in a horizontal configuration that was partially prefilled with oil. We emphasize the prefilling liquid's role in the dynamics of the system and show the influence of the displacing liquid's viscoelastic properties. We find significant changes in the capillary regime as the displacing liquid's viscoelastic properties change.*

Key Words: *capillary flow, capillary displacement, viscoelastic fluids, immiscible liquids*

1. INTRODUCTION

At spatial scales smaller than the capillary length, forces due to surface tension become dominant and liquids begin to flow despite other acting forces such as gravity. When a liquid comes into contact with a tube of radius smaller than the capillary length, it will start to invade the tube [1]. As capillary flows are natural and spontaneous phenomena and do not require any external additional forces, they have been implemented in many applications of various fields. They present advantageous solutions to technical problems, such as capillary pumping systems for solar heating applications [2], cooling systems for fuel cells [3] and vapor generation [4].

The dynamics and regimes of capillary flows have been extensively described in past papers [5-7]. Capillary invasion of non-Newtonian liquids have been investigated in both horizontal and vertical alignments [8, 9] and theoretical models have been validated experimentally [10].

The capillarity-driven displacement of liquid slugs in horizontal tubes has been approached both experimentally and theoretically [11-13]. The capillary flow in partially prefilled tubes has been previously investigated using a system of Newtonian immiscible liquids [14]. Studies regarding the spontaneous flows of a viscoelastic fluid in a capillary tube partially filled with another immiscible liquid have been conducted in few previous studies.

The paper is concerned with the capillary displacement of sunflower oil when a partially filled tube touches the free surface of a viscoelastic liquid. The response of the system, with respect to the viscoelastic properties of the liquid, is investigated using polyethylene oxide solutions. The dynamics of the two-liquid system is also emphasized by varying the pre-filling liquid's volume. The paper is structured as follows: first, in section 2, the material properties of the working liquids are given followed by the experimental details concerning the method that was used to capture the relevant dynamics. Section 3 is dedicated to the study of a single viscoelastic liquid phase invading the capillary tube. In section 4 we show the effect of a prefilling Newtonian oil on the advance of the two immiscible liquid system.

2. EXPERIMENTAL DETAILS

A schematic representation of the experimental setup is shown in figure 1-a). The experiments are performed in two scenarios, the viscoelastic solution advancing in an empty or partially prefilled capillary tube with an immiscible liquid. In the case of a single working liquid, a glass capillary tube with an inner diameter of 0.95 mm and a length of 50 mm is placed horizontally and put in contact with liquid bath containing a viscoelastic fluid. For the two-liquid system, a horizontally positioned capillary tube is first brought in contact with a Newtonian oil to create a column of liquid, partially pre-filling the tube. By adjusting the contact time, different column lengths are obtained. The partially filled capillary tube is then brought to the free surface of a viscoelastic fluid contained in the liquid bath.

For both cases, as the viscoelastic fluid starts invading the capillary, a high-speed camera working at 500 fps is used to record the advance of the meniscus. The resulting frames are analysed with an in-house image-processing routine implemented in MATLAB, which detects and calculates the position of the advancing meniscus in the capillary tube. Representative visualizations of both cases of capillary flow are displayed in figure 1-b).

Viscoelastic polyethylene oxide solutions of varying concentrations are used as the invading fluid. The experiments were carried out using the 1000 ppm (0.1g/100ml), 10000 ppm (1g/100ml), 20000 ppm (2g/100ml) and 40000 ppm (4g/100ml) concentrations. Their respective shear stress as a function of shear rate and the corresponding viscosity curves are presented in figure 2 and show a shear-thinning behavior. Experiments were also performed with water of density $\rho = 998\ \mathrm{kg/m^3}$, viscosity $\eta = 1\ \mathrm{mPa \cdot s}$, and surface tension $\gamma =$

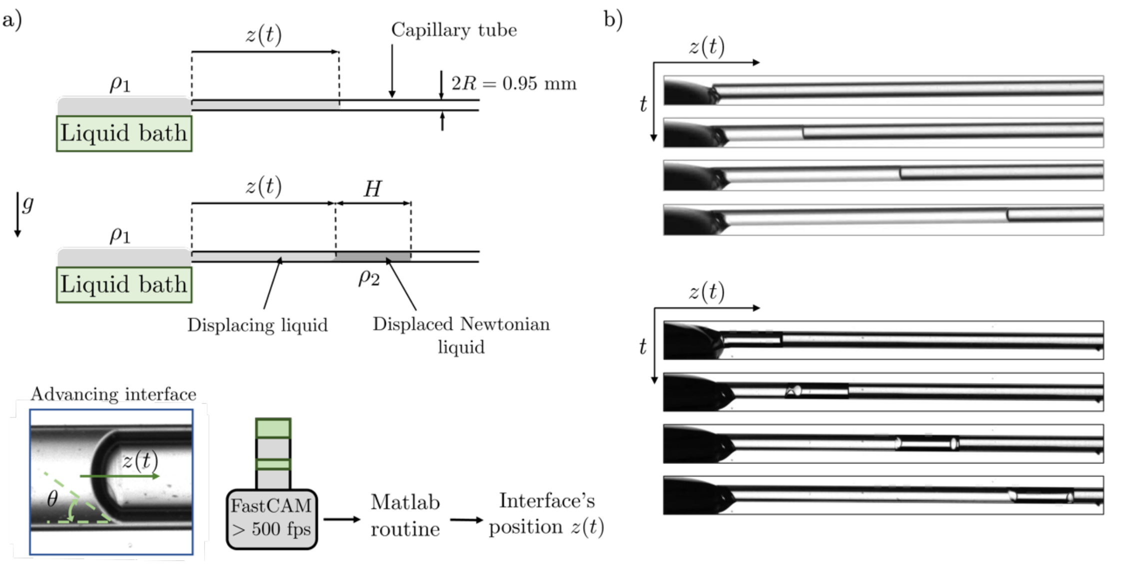

Fig. 1 – a) Details of the experimental setup; b) Capillary flow in horizontal tubes with (lower set of images) and without a prefilling liquid column

72 $m\text{N/m}$ as the displacing liquid in order to compare the dynamics of the fluids with weakly viscoelastic solutions. In the case of the two-liquid system, the capillary tube is partially prefilled with a column of sunflower-seed oil of density $\rho = 920\,\text{kg/m}^3$, viscosity $\eta = 55\,\text{mPa}\cdot\text{s}$ and surface tension $\gamma = 24\,m\text{N/m}$.

The properties of the working liquids were determined using standard procedures. The densities of the fluids are obtained by the mass-per-volume method. A rotational rheometer (Anton Paar Physica) with a cone-plate geometry was used to measure the viscosities of the Newtonian working liquids and to obtain the shear stress as a function of shear rate and the corresponding viscosity curves of the viscoelastic PEO solutions (see figure 2). The surface tensions are determined via the pendant drop method, by measuring two diameters within the suspended droplet at two selected planes, the equatorial plane and the plane at a distance equal to the equatorial diameter from the tip of the drop.

3. CAPILLARY FLOW OF VISCOELASTIC PEO SOLUTIONS

At the contact between the free surface of a liquid and a capillary tube, the fluid will naturally advance due to dominant surface tension forces. For the case of a single working liquid, the capillary invasion of polyethylene oxide solutions is analysed. As a starting point, pure water is put in contact with the glass capillary tube, its dynamics representing a benchmark problem. The position of the water-air interface with respect to time is depicted in figure 3-a). A typical Washburn flow can be observed as the position of the water-air interface could be described by a power law function of time. Being a fluid with low viscosity and high surface tension, water quickly reaches distances comparable with the length of the capillary tube (approximately 43 mm in almost 0.33 s). The initial high velocity slightly decreases in time due to increasing viscous friction forces. The same observations can be noted for the weakly viscoelastic substance used, the aqueous PEO solution of 1000 ppm concentration. Due to its

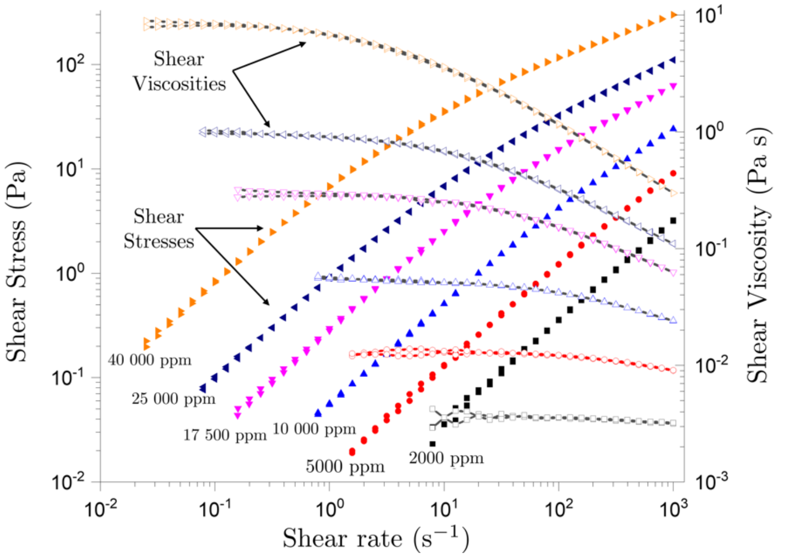

Fig. 2– Rheological measurements for the working viscoelastic fluids (polyethylene oxide solutions), shear stress versus shear rate (left vertical axis) and shear viscosity versus shear rate (right vertical axis). For the viscosity curves the concentration of PEO decreases from top to bottom as depicted for the corresponding shear stress from left to right

low concentration, similar behaviour to water is observed. The liquid invades the capillary tube in a short time frame, reaching the same distance as water in approximately 0.36 s. The meniscus' position in respect to time of the weakly viscoelastic PEO substance is depicted in figure 3-a). The results are presented comparatively with the experimental data obtained for water and small differences between the dynamics of the two liquids can be observed. The experimental data still suggest a Washburn type flow, but the capillary invasion is slightly delayed due to the addition of viscoelasticity.

By increasing the polymer concentration 10 or 20 times, the dynamics of the phenomena indicate the same type of flow, but significant differences regarding the time scales occur. The dynamics of resulting solutions (PEO of 10 000 ppm and 20 000 ppm concentrations) are presented comparatively in figure 3-b). It can be noted that, in order to reach the same position in the capillary tube, the two substances require different time scales. As seen in figure 3-b), in less than 2 seconds the smaller concentration of polymer solution invades almost half of the capillary tube (approximately 27 mm). By doubling the concentration, the invasion of the capillary tube is drastically delayed, as the meniscus requires more than 32 seconds in order to reach the same spatial position.

The advance of the interface's position in time for all invading liquids, pure water and the viscoelastic PEO solutions, is depicted in figure 4-a). Significant differences between the time scales of each experiment can be seen. It can be easily observed that by adding and increasing the viscoelastic component, the dynamics of the capillary invasion is delayed. However, regardless of the viscous and elastic behaviors, the advance of the meniscus in time in the tube follows a similar type of power law.

Time scale differences can be further emphasized by analyzing and comparing the time taken by each of the working liquids in order to reach the same distance in the capillary tube. The effect of the increased concentration of PEO on the time scale of the capillary flow is highlighted in figure 4-b). For an advance of less than 10 mm, the necessary time scale varies from approximately 0.06 s (for the weakly viscoelastic liquid) to almost 3 minutes (for the highest concentrations of polymer, 40 000 ppm). The viscoelastic solutions require different periods of time in order to fill (or partially fill) the capillary tube.

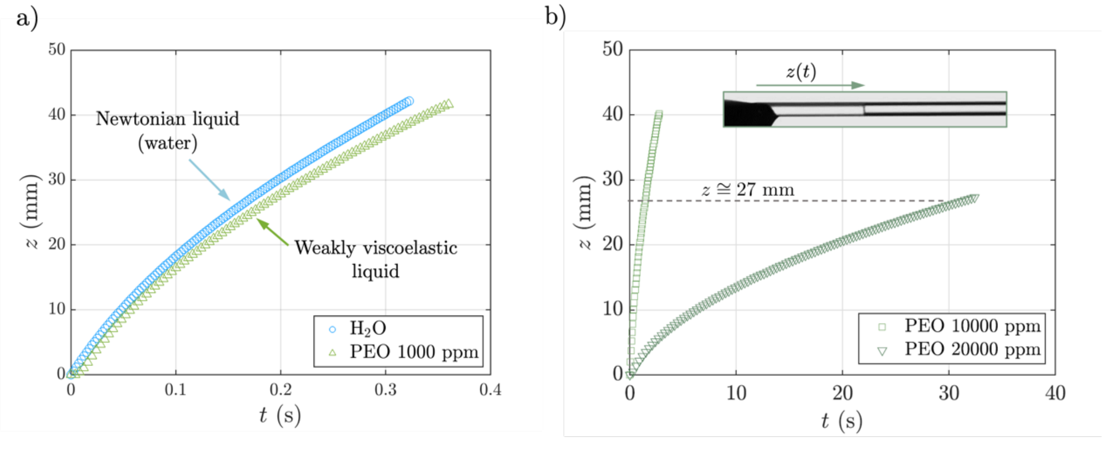

Fig. 3 - Comparative experimental results of the interface's position in time for: a) pure water and the lowest concentration of PEO, 1000 ppm; b) two viscoelastic solutions of different concentrations, 10 000 and 20 000 ppm

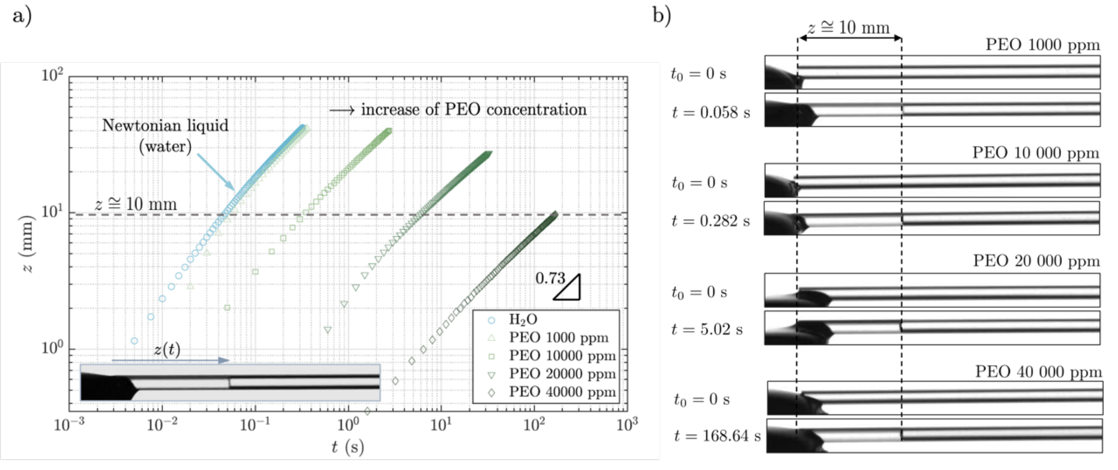

Fig. 4 – a) Logarithmic representation of the meniscus dynamics for water and the polyethylene oxide solutions; b) Series of images and data showing the viscoelastic component's effect on the time scale of the capillary flow.

4. DISPLACING A NEWTONIAN LIQUID USING VISCOELASTIC PEO SOLUTIONS

The capillary tube is first brought into contact with the free surface of Newtonian sunflower oil. The contact time will then give the prefilling liquid's column length inside the tube. In this manner, an oil slug is created and its volume can be adjusted by means of the contact time. The now partially filled capillary is brought into contact with the free surface of the viscoelastic polyethylene oxide solutions. The non-Newtonian liquid starts flowing in the tube generating the displacement of the oil column. The experiments are conducted with at least three different lengths of the oil slug for each concentration of the PEO solutions. The positions of the interface between the Newtonian oil and the viscoelastic liquid with respect to time will be depicted.

For a weakly viscoelastic fluid as the invading liquid (PEO 1000 ppm), the displacements of different oil column lengths in time are presented in figure 5-a). The dynamics of the capillary flow with three oil column lengths are compared with the meniscus advance in the case of no prefilling liquid. The addition of an immiscible liquid of column length H results in a linear variation of the interface's position with respect to time.

This variation indicates that the two liquid system advances with constant velocity. For short length scales comparable to the capillary tube's length and for low polymer concentrations of the displacing liquids, the constant viscous friction effects of the oil column are dominant and result in a steady capillary invasion.

It can be observed that the presence of an immiscible liquid strongly delays the capillary invasion. As the oil column's length increases, the flow velocity decreases but remains constant in time.

By using a viscoelastic PEO solution of higher concentration, the quasilinear variation can no longer be observed. We increase the polymer concentration of the previous working liquid tenfold and observe the dynamics in the case of four different oil column lengths. The advance of the two-liquid system in time is presented in figure 5-b). The addition of an immiscible prefilling liquid and the increase of its volume cause an obvious setback to the dynamics of the capillary flow. The quasilinear variation of the interface's position in time is

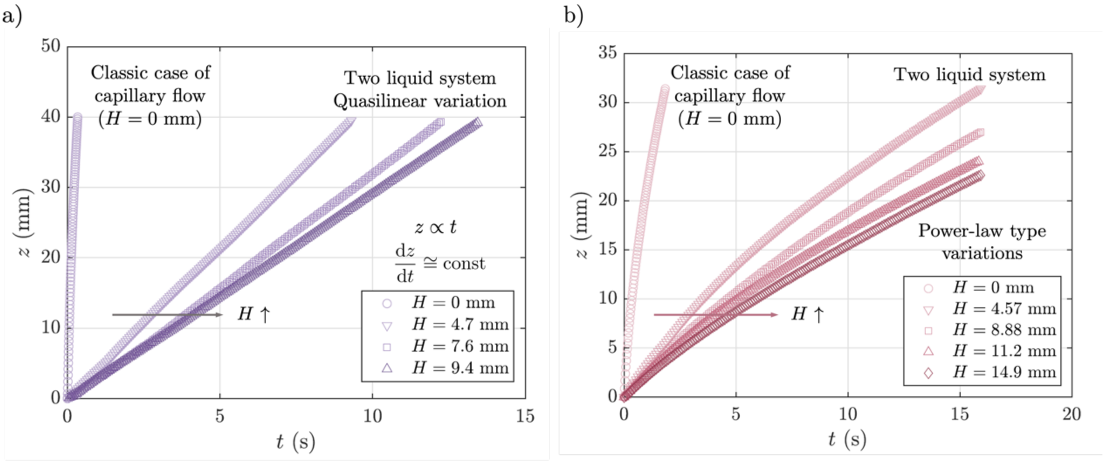

Fig. 5 – The liquid-liquid interface's position with respect to time for different oil column length for the displacing PEO solutions of concentrations: a) 1000 ppm; b) 10 000 ppm

no longer observed and the liquid system's velocity can no longer be considered constant even for short length scales.

Due to the more pronounced viscoelastic behaviour of the higher concentration, the dynamics start to resemble a Washburn type flow. The viscous friction effects given by the non-Newtonian liquid and the oil column are comparable. The increase of polymer concentration and of the displaced liquid volume lead to a significant delay in the capillary invasion.

We generate an approximately equal oil column of length H for all of the working liquids, water and viscoelastic PEO solutions. The interface's position in time, $z(t)$, is investigated as a function of the polymer concentration.

The experimental data for $H \cong 4.3$ mm are presented in figure 6. The most rapid capillary invasion is exhibited by pure water as a displacing liquid, with a high constant flow velocity. The flow of the weakly viscoelastic fluid, PEO 1000 ppm solution, is slightly delayed due to the addition of viscoelasticity.

For distances comparable to the capillary tube's length, the liquid-liquid interfaces of water and the weakly viscoelastic solutions present a quasilinear variation with respect to time. Increasing the polymer concentration, the viscoelastic properties deflect the dynamics of the two-liquid system from a seemingly constant velocity to a typical Washburn flow and the capillary flow is delayed.

Conducted experiments reveal a spontaneous phenomenon of droplet generation. As the viscoelastic fluid displaces the oil column, the deposited oil film undergoes a capillary instability.

The meniscus separating the two immiscible liquids is initially concave. When instability occurs, a breakup of the interface occurs and a droplet of polymer solution is formed.

The drop flows through the displaced oil column and the meniscus regains a concave shape throughout the capillary invasion. The phenomenon was first observed in the case of water-oil systems [14].

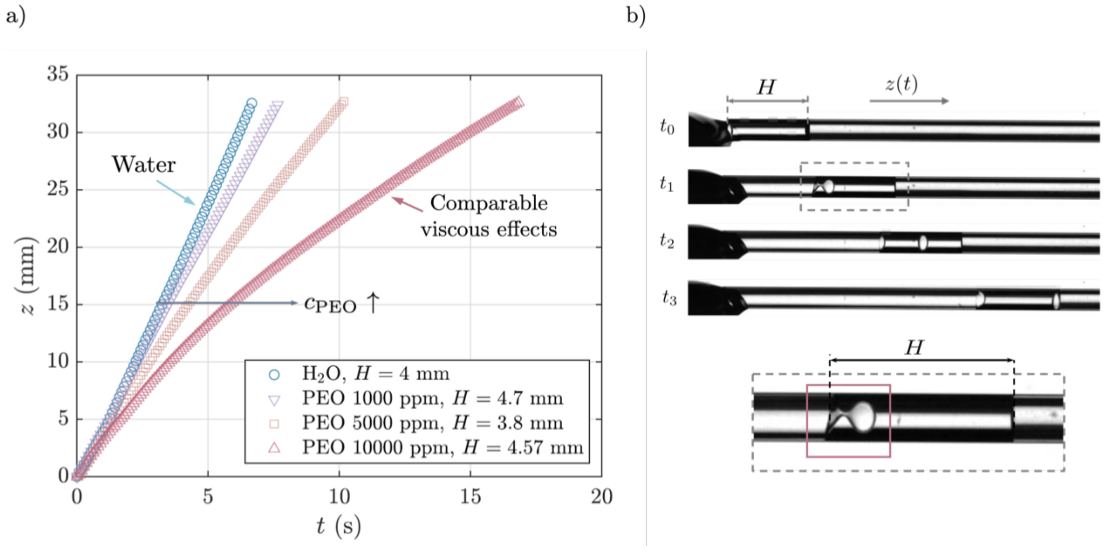

Fig. 6 – a) Capillary displacement of an approximately equal volume of oil, $H \cong 4.3$ mm, by water and various concentrations of the PEO solutions; b) Series of images depicting the capillary displacement of an oil column with detail on the droplet generation phenomenon

5. CONCLUSIONS

The paper tackles the problem of non-Newtonian capillary flow in classic and partially prefilled tubes. We present experimental data concerning the flow of viscoelastic PEO solutions in the two cases. In the two-liquid system scenario, the tube was prefilled with a Newtonian oil. We emphasize the influence of the volume of oil contained in the tube prior to the invasion of the viscoelastic PEO solution and the influence of the polymer concentration.

The dynamics of the PEO solution into an empty capillary tube suggest a Washburn-type flow. The data for the weakly viscoelastic liquid are similar to the results obtained for pure water. The two fluids exhibit a quick invasion of the tube due to high surface tensions and low viscosities. The addition of viscoelasticity causes a slight delay in the liquid's invasion. By increasing the polymer concentration, the viscoelastic properties of the substance lead to an obvious setback of the capillary flow. In the two-liquid system, the same remark is noted. For equal volumes of the prefilling liquid, the invasion of the displacing liquid is delayed if the polymer concentration is increased. The displaced liquid's volume also influences the phenomenon. Using the same concentration of PEO and increasing the oil column's length, the opposing viscous forces are increased. The capillary flow of the two-liquid system is then delayed.

For distances comparable to the capillary tube's length, weakly viscoelastic solutions in tandem with a column of prefilling immiscible liquid flow with constant velocity. The viscous forces of the sunflower oil column are dominant to those exhibited by the non-Newtonian liquid. This is no longer valid when the viscous and elastic components are increased due to an increase of polymer concentration and the viscous friction effects of the two flowing liquids become comparable.

Experiments show a natural phenomenon of droplet formation and detachment at the interface between the viscoelastic fluid and Newtonian liquid. The phenomenon was first reported in the case of Newtonian liquid systems.

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