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Displacement flows of dilute polymer solutions in capillaries

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Abstract

The two-dimensional, free surface flow of a viscoelastic liquid being displaced in a capillary tube by gas injection is analyzed by both experiments and theory. The experiments consisted of flow visualization and measurement of mass displaced by a gas bubble in a capillary tube filled with a viscoelastic liquids. Various solutions of low molecular weight polyethylene glycol (PEG) and high molecular weight polyethylene oxide (PEO) in water were used in order to evaluate the effect of viscoelastic behavior on the flow. The rheological properties of the solutions were evaluated both in shear and predominantly extensional flows. The theoretical analysis consisted of solving the conservation equations, together with appropriate constitutive models, with the finite element method. Models of flows of high molecular weight polymer solution must rely on theories that can account for the different behavior of microstructured liquids in shear and extensional flow. Moreover, displacement flows involve a free surface, and the domain where the differential equations are posed is unknown a priori, being part of the solution. These two characteristics make the problem extremely complex. Here, the viscoelastic behavior of the liquid was described using three differential constitutive equations that approximate dilute and semi-dilute polymer solutions, namely Oldroyd-B, FENE-P and FENE-CR . Both the theoretical predictions and experimental measurements were obtained at values of the elasto-capillary number much higher than previously reported. At this limit, a new phenomenon is observed, the film thickness attached to the capillary wall reaches a plateau as the liquid becomes more elastic.

Keywords: Free surface flow; Viscoelastic flow; Gas-assisted injection molding; Gas–liquid interface

1. Introduction

The displacement of a liquid inside small passages and capillary tubes by another liquid or gas occurs in many practical situations. Important examples include the flow inside the porous space of a reservoir in enhanced oil recovery methods, coating process of catalytic converter and inside tubes, and gas-assisted injection molding. These flows belong to a class of flows generally referred to as free surface flows; the configuration and position of the interface between the two fluids is unknown a priori and is part of the solution of the problem.

Fig. 1 shows the region close to the tip of the interface of a liquid inside a capillary tube being displaced by a gas bubble moving at speed U. It is more convenient to work in a frame in which the bubble is stationary and the capillary wall moves at a speed U in the opposite direction, as illustrated. The more liquid is left on the wall then the less efficient is the displacement process. This information is one of the main goals of theoretical and experimental analysis of displacement flows.

A dimensionless measurement of the thickness of the liquid film attached to the wall, $t = R_0 - R_b$, used in the literature is the fractional coverage m, defined as the fraction of the tube cross-sectional area coated with liquid after the bubble penetration, i.e.

$$m = 1 - \frac{R_b^2}{R_0^2}. \tag{1}$$

R_0 is the tube radius and R_b is the radius of the penetrating bubble. Fairbrother and Stubbs [11] were the first to experimentally study the displacement of a Newtonian liquid in a capillary tube by a gas. They found that the fractional coverage m was a function of the capillary number $Ca \equiv \eta U/\sigma$. η is the viscosity of the liquid, U is the bubble velocity, σ is the interfacial tension between the inviscid penetrating gas and the displaced liquid. Their data were well fitted by a power-law relation in the range of $10^{-3} < Ca < 10^{-2}$:

$$m \approx Ca^{1/2}.$$

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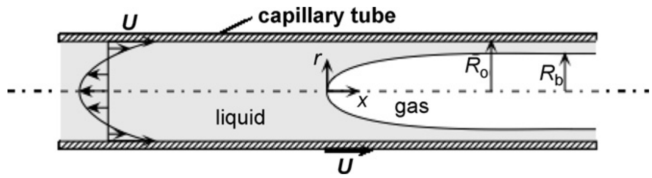

Fig. 1. Sketch of a liquid-displacement problem.

The first theoretical analysis of this problem was presented by Bretherton [3]. The analysis was focused on the frontal and transition regions of the gas bubble. He assumed zero shear stress at the interface and small capillary numbers. A relation between the fractional coverage m and the capillary number was derived:

$$m = 1.29(3\text{Ca})^{2/3}.$$

Taylor [26] extended the range of capillary number in the experiments of liquid displacement in capillaries to $10^{-3} < \text{Ca} < 1.9$. He found that the fractional coverage asymptotically approached $m = 0.56$ as the capillary number tends to $\text{Ca} = 2$.

Theoretical predictions of this flow at larger values of capillary number are only possible with the full solution of the problem, that can only be computed numerically. Reinelt and Saffman [21] predicted the fractional coverage as a function of capillary number for $10^{-2} < \text{Ca} < 2$ for both planar and axisymmetric flows. They used finite difference method to discretize the governing equations. Their results are in good agreement with experiments reported in the literature. Giavedoni and Saita [13], using finite element method, extended the range of capillary number in the analysis to $10^{-5} < \text{Ca} < 10$. Their results also agreed well with experiments.

The literature on displacement of viscoelastic liquids in capillaries and Helle-Shaw cells by a gas bubble is more recent and scarce. Ro and Homsy [22] presented a theoretical analysis of the effect of liquid elasticity on the meniscus shape and film thickness left on the wall when a bubble displaces a polymeric liquid in a Helle-Shaw cell. They used perturbation expansions on capillary and Weissenberg numbers to solve the problem, i.e. their predictions were restricted to small values of both dimensionless numbers. The Weissenberg number was defined as $\text{Wi} \equiv \lambda\dot{\gamma}_\text{w}$. λ is the relaxation time of the liquid and $\dot{\gamma}_\text{w}$ is the (Newtonian) shear rate at the wall, assumed to be characteristic of the process. The viscoelastic behavior of the liquid was described by the Oldroyd-B constitutive model. The main conclusion was that, as the liquid becomes more elastic, the film thickness deposited on the wall is slightly reduced due to resistance to streamwise strain. Huzyak and Koelling [16] investigated experimentally the penetration of a long gas bubble through a tube filled with a viscoelastic liquid. They used two Boger liquids (viscoelastic liquids with constant shear viscosity) and reported the fractional coverage m as a function of the capillary number and Weissenberg number. The results show strong thickening of the liquid layer deposited on the tube wall relative to Newtonian results as the liquid becomes more viscoelastic. The extensionally thickening viscosity of the polymeric solution at the extension dominated flow near the bubble tip was pointed as a possible cause for the increase on the film thickness deposited on the wall as the Weissenberg number rises. This hypothesis was confirmed by Gauri and Koelling [12], who measured the velocity field around the bubble tip using PTV. In their experiments, the highest elasto-capillary number, defined as the ratio of the Weissenberg to capillary number, explored was close to $\text{Wi}/\text{Ca} \approx 20$.

Full solution of viscoelastic free surface flows is quite challenging. Only recently this type of problem has been addressed with success. Pasquali and Scriven [19] and Lee et al. [18] have shown that elastic stress boundary layers attached to the free surface are formed when the kinematics close to that region is dominated by extensional deformation. The change in the forces acting near the free surface affects the flow in that region and consequently the meniscus configuration. Lee et al. [18] analyzed the effect of viscoelasticity on the displacement of a polymeric liquid inside a Hele-Shaw cell by a long gas bubble and on the downstream portion of a slot coating flow. The viscoelastic behavior of the liquid was modelled with the Oldroyd-B, FENE-P and FENE-CR constitutive equations. The finite element method was used to discretize the governing equations. They predicted the same qualitative trend observed by Huzyak and Koelling [16], and Gauri and Koelling [12], however the predictions were limited to values of elasto-capillary number below $\text{Wi}/\text{Ca} \approx 7$.

In this paper, the displacement of a polymeric solution inside a capillary tube by a long gas bubble is examined by experiments and theory. The gas–liquid interface and the film thickness deposited on the capillary wall were visualized for different solutions of high molecular weight polymer (PEO) and low molecular weight (PEG) in water. The test liquids were designed so that their shear viscosity was constant, i.e. Boger liquid behavior. The level of the elastic response of the liquid was varied by using different molecular weights of the PEO, from $M_\text{w} = 0.9 \times 10^6$ to $M_\text{w} = 8 \times 10^6$ g/mol. The test liquids extends the range of the elasto-capillary number explored by Huzyak and Koelling [16], and Gauri and Koelling [12]; experiments were performed at $\text{Wi}/\text{Ca} \approx 92$. The theoretical analysis consisted of solving the conservation of momentum and continuity equations for steady, two-dimensional flow with free surfaces, coupled with differential constitutive models that describe the mechanical behavior of polymer solutions. Three models for dilute and semi-dilute polymer solutions were tested: Oldroyd-B, FENE-CR, and FENE-P constitutive relations. The system of equations was solved by the Galerkin and Petrov-Galerkin/finite element method. The mechanisms responsible for the variation of the thickness of the liquid layer deposited on the capillary wall as a function of the rheological properties of the liquid are discussed in terms of the stress field components along the streamlines. The predictions were obtained at values of the elasto-capillary number up to $\text{Wi}/\text{Ca} \approx 50$, also extending the range previously reported in the literature.

2. Theoretical analysis

2.1. Mathematical formulation

The flow in the region close to the gas bubble tip was described by the complete two-dimensional, steady-state mass