



Rising bubble instabilities and fragmentation in a confined polymer solution



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ABSTRACT

This work investigates the dynamics of a single bubble rising in a polymer solution confined in a vertical Hele-Shaw cell. Different mixtures of PEO (polyethylene oxide) in water have been used, which exhibit a Newtonian viscosity plateau, followed by a shear-thinning behavior above a shear rate of 0.1 to 1, typically. Depending on the bubble volume, different regimes are reported. When increasing the bubble volume, a transition is observed from a small, round bubble to a cusped bubble, which exhibits a singularity at the rear. Below a critical volume, the bubble rises vertically and does not exhibit any apparent shape deformation. However, above a critical volume, the cusped bubble develops a peculiar instability. Its front flattens at a given angle respect to the horizontal, leading to either a deflection in its trajectory, or its fragmentation. We characterize these two dynamics and interpret the fragmentation process in terms of a viscous fingering recalling the Saffman–Taylor instability. Interestingly, the finger growth is directly controlled by the bubble size.

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1. Introduction

Bubbles rising in non-Newtonian fluids exhibit a wide range of puzzling behaviors which have excited different research communities for decades. Indeed, they are widely encountered in nature, from degassing on the ocean floor [1,2] to giant bubbles rising and bursting on volcanoes [3–6]. In both cases, the complex nature of the surrounding fluid (soft immersed sediments, bubble- or crystal-rich magma [1,7–9]) give rise to non-Newtonian effects, such as shear-thinning behavior [9,10]. This effect strongly couples to the bubble rise dynamics and often makes it difficult to interpret the field data. From a more fundamental point of view, experimental studies of bubbles rising in non-Newtonian fluids have started in the 60's, leading to a large amount of either experimental or numerical works which strongly developed since the 80's [11]. Among the many peculiar observations are the deformation of the bubble shape respect to Newtonian fluids [12–14], the presence of a singularity at the bubble rear (cusp) [11,15] and a negative wake due to elastic properties [16], oscillations of the bubble shape or velocity [17,18], or the existence of a discontinuity in the bubble terminal velocity for a critical volume [19,20]. All these phenomena have been reported for bubbles rising in unconfined non-Newtonian fluids.

On the other hand, the influence of fluid confinement can have drastic consequences on the bubbles dynamics and, more generally, on the gas invasion dynamics in viscous fluids. The pioneer study is the classical Saffman–Taylor problem, consisting in the injection of air at constant pressure in a horizontal quasi two-dimensional cell (Hele-Shaw cell) filled with a Newtonian, viscous fluid [21,22]. It has been shown that the initially planar interface deforms, leading to the growth of a finger-like instability, which has been widely studied theoretically [23] and experimentally [24]. More recent studies considered the Saffman–Taylor instability in the case of non-Newtonian fluids. They pointed out that for weakly shear-thinning fluids (dilute polymer solutions), using an effective Darcy's law with the shear-thinning viscosity accounts for the finger width [25]. For strong shear-thinning fluids, narrower fingers are reported [25]. When the fluid exhibits normal stress effects rather than shear-thinning effects, however, the experiments show a finger widening [26,27].

Although apparently simple, the problem of a single bubble rising under its own buoyancy in a confined, non-Newtonian fluid has not been tackled yet. Even the simpler case of a single bubble rising in a viscous, Newtonian fluid such as water, mentioned by Taylor and Saffman since 1959 [28], is still investigated nowadays [29–31]. In particular, most studies focus on the triggering of path instabilities, an oscillatory bubble motion observed either in confined or unconfined cell, and which triggering depends on the fluid rheology [32–35].

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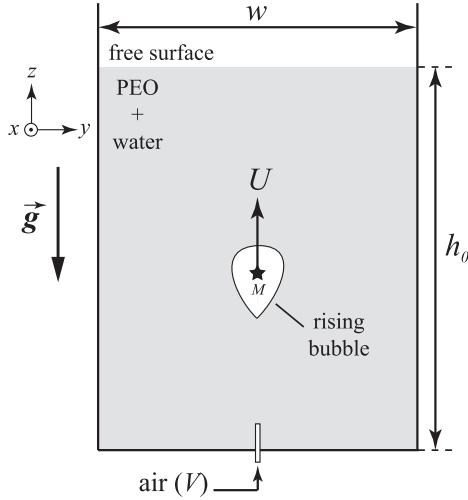


Fig. 1. Sketch of the experimental setup. A single bubble (volume V) is injected at the bottom of a vertical Hele-Shaw cell of width w , filled with a PEO + water solution up to a height h_0 . It rises with a vertical velocity U . Its center of mass is M [black star, coordinates (y_M, z_M)].

Here, we present the experimental study of a single bubble rising in a vertical Hele-Shaw cell. The surrounding fluid is a solution of long polymer chains (see Section 2.2), characterized by a strong shear-thinning behavior. The polymer concentration is chosen high enough so that (1) non-Newtonian effects are significant and (2) the local viscosity is always high enough to prevent the formation of the classical path instabilities. We report a new type of instability (Section 3), leading to either the bubble horizontal deflection or fragmentation.

2. Experimental setup

2.1. Description

The experimental setup consists of a Hele-Shaw cell made of two glass plates (width $w = 28.3$ cm, height 40 cm, gap $e = 2$ mm), which are systematically removed and carefully cleaned with water, ethanol and optical paper before each series of experiments. The cell is then filled with a polymer solution (see Section 2.2) up to a height $h_0 \approx 30$ cm (Fig. 1). Air is injected manually through a nozzle (inner diameter 1 mm) by means of a syringe (capacity 60 mL). By tuning the amplitude and velocity of the injection, the volume V of the single injected bubble can be varied, typically between 0.01 and 1.5 mL (see Section 2.3). After the injection, the bubble rises by buoyancy. We denote U its vertical velocity and M its center of mass [coordinates (y_M, z_M)] (Fig. 1). The origin of the coordinates $(0, 0)$ is taken at the injection nozzle, at the bottom center of the cell.

A homogeneous backlight is ensured by a transparency flat viewer (Just NormLicht, Classic Line) located behind the cell. Direct visualization of the bubble dynamics is performed using a video camera (Pixelink PL-B781) with an adjustable lens (12.5–75 mm with a focal distance of 1.2 m), a resolution of 1280×416 pixels and a frame rate of 41 fps.

2.2. Fluids characterization

The fluids used in these experiments are polymer solutions of polyethylene oxide (PEO) in water. The PEO is sold as a white powder which can be diluted in water at different concentrations, making it possible to tune the rheological properties of the mixture [PEO+water]. It is characterized by a high molecular weight, $M_w =$

Table 1

Parameters of the Carreau–Yasuda law fitting the rheology of the four PEO solutions (see gray lines, Fig. 2).

c [g/L]	η_0 [Pa s]	τ [s]	n [-]	a [-]
3	0.12	0.69	0.59	2.76
4	0.49	1.92	0.52	2.04
5	1.37	4.19	0.48	2.02
6	4.12	6.09	0.40	1.12

8×10^6 g/mol (Sigma-Aldrich, 372838). For polymer solutions, one can define the entanglement concentration of the polymer chains as

$$C^* = \frac{M_w}{(4/3)\pi R_g^3 \mathcal{N}_a}, \quad (1)$$

where M_w is the molecular weight, R_g the gyration radius of the polymer and \mathcal{N}_a the Avogadro number [36]. For the PEO described above, the entanglement concentration is $C^* = 0.04$ g/L. Four different polymer solutions were prepared with the following concentrations: $c = [75, 100, 125, 150]C^*$, corresponding to $c = [3, 4, 5, 6]$ g/L. These high concentrations ensure that (1) the polymeric chains are well-entangled, (2) the viscosity is higher than the water viscosity and (3) the non-Newtonian effects will be non-negligible in our experiment. The value of the surface tension has been measured using a commercial tensiometer (Tracker, Teclis) based on the rising bubble technique, equivalent to the pendant drop [37,38]. For the solution at $c = 100C^*$, the surface tension is $\sigma = 64 \pm 2$ mN/m. It is assumed to be the same for all the polymer solutions.

The rheological properties of the polymer solutions have been characterized by a rheometer (Bohlin C-VOR 150) with a cone-plane geometry (diameter 60 mm, angle 2°). Fig. 2a displays the fluids dynamic viscosity η as a function of the applied shear rate $\dot{\gamma}$. The four polymer solutions exhibit a viscosity between about 10–1000 times the viscosity of water, and a shear-thinning behavior above a critical shear rate $\dot{\gamma}_c$, in agreement with previous studies on PEO solutions rheology [39]. Note that $\dot{\gamma}_c \approx 0.1$ – 1 s $^{-1}$, of the order of magnitude of the shear rates in our experiments.

The shear-thinning behavior of entangled flexible polymers has been often modeled by the Carreau law, which takes into account both the Newtonian plateau at low shear rate and the decreasing slope at higher shear-rate [40]. However, we observe slight differences in the decreasing slope for the different concentrations, which is not compatible with the power $-1/2$ predicted by this simple law. Here, we use the more generalized Carreau–Yasuda law [41]

$$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{[1 + (\tau \dot{\gamma})^a]^{\frac{1-n}{a}}}, \quad (2)$$

where η_0 is the viscosity at zero shear rate, η_∞ the viscosity at infinite shear rate (or solvent viscosity), τ a characteristic time, n a dimensionless exponent and a a dimensionless parameter describing the transition between the first Newtonian plateau and the shear-thinning region [41]. For all the solutions, the solvent is water and $\eta_\infty = 10^{-3}$ Pa s. Table 1 summarizes the fitting parameters (η_0 , τ , n , a) for the PEO solutions of different concentrations (see gray lines in Fig. 2).

Fig. 2b displays the first normal stress coefficient, $\psi_1 = N_1/\dot{\gamma}^2$, where N_1 is the first normal stress difference for the four PEO solutions. For PEO solutions, the first normal stress difference is predicted to increase quadratically with the shear rate [42]. However, a less-than-quadratic behavior is observed with ψ_1 decreasing with $\dot{\gamma}$, as already reported for PEO-water mixtures and within the same order of magnitude than in the literature [39].

The viscoelastic properties of the fluids have been measured with a rheometer TA Instruments, AR100 with a cone-plate geometry of diameter 40 mm, angle 2° . Fig. 2c displays the elastic

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