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Self-oscillations of a cusped bubble rising through a micellar solution

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Abstract Observations of the motion of a rising bubble in a wormlike micellar system are presented, obtained in both a rotating toroidal cell and a stationary box. For equimolar concentrations of aqueous CTAB/NaSal (8–11 mM), bubbles larger than a certain volume undergo oscillations in shape which include an

apparent cusp. The average frequency of these oscillations is found to increase with bubble volume.

Key words Bubble · Cusp · Oscillating instabilities · Viscoelastic surfactant solution

Introduction

There are several fascinating phenomena observed when a bubble rises through a non-Newtonian or viscoelastic fluid, one of which is an apparent cusp at the trailing end of the bubble (Chhabra and De Kee 1992; Chhabra 1993). This occurrence was first reported by Philippoff (1937), and since then many experimental studies have reported some or all of the following effects: a cusp-like tail (Astarita and Apuzzo 1965; Hassager 1979; Coutanceau and Hajjam 1982; Bird et al. 1987; Liu et al. 1995; Rodrigue and De Kee 1999), a discontinuity in the terminal velocity as a function of volume (Chhabra 1993; Astarita and Apuzzo 1965; Liu et al. 1995; Rodrigue and De Kee 1999; Rodrigue et al. 1996; Leal et al. 1971; Zana and Leal 1978; Acharya et al. 1977), and a “negative wake” in which the fluid flows away from the bubble (Bird et al. 1987; Hassager 1979; Bisgaard 1983); see (Chhabra and De Kee 1992; Chhabra 1993) for more complete reviews. It is still not clear how these effects are connected to each other, or which viscoelastic properties are responsible for which effects (Liu et al. 1995). For instance, cusped bubbles are observed in purely elastic (Boger) fluids (Coutanceau and Hajjam 1982; Liu et al. 1995), and in shear thinning viscoelastic fluids (Coutanceau and Hajjam 1982). In this paper we present observations of cusped bubbles in

another kind of fluid, an aqueous solution of the micellar system cetyltrimethylammonium bromide (CTAB)/sodium salicylate (NaSal) (Rehage and Hoffmann 1982; Cates and Candau 1990; Hu et al. 1993). Unlike other non-Newtonian fluids, the cusp is not steady but oscillates in time as the bubble rises. To our knowledge, rising bubbles have not been studied in this system.

The shape and motion of a gas bubble rising through a fluid is an important example of a free boundary problem (Stone 1994; Davies and Taylor 1950), for which the behavior is very different in Newtonian and non-Newtonian fluids (Bird et al. 1987). No cusp is observed when a bubble rises in a Newtonian fluid (Coutanceau and Hajjam 1982; Bhaga and Weber 1981), unless it rises along an inclined wall (Liu et al. 1995). Although cusps at fluid interfaces are usually only encountered in calculations where surface tension has been neglected, apparent cusps are observed in real fluids at low Reynolds numbers (Joseph et al. 1991; Jeong and Moffatt 1992), for which the calculated radius of curvature due to surface tension is surprisingly small (Jeong and Moffatt 1992). The transition of a rising bubble to a cusped shape is known to occur above a certain critical bubble volume (Chhabra 1993), at which a jump in the velocity-volume relation occurs (if it occurs at all). This jump has been associated with the transition

from no-slip to free shear conditions at the bubble interface (the Hadamard-Rybczynski transition) (Astarita and Apuzzo 1965; Leal et al. 1971), though there is some disagreement with the details of this hypothesis (Rodrigue and De Kee 1999). Another explanation of the jump is that it results from a sudden drag reduction due to the change in bubble shape at the cusp instability (Liu et al. 1995). The question remains why a non-Newtonian fluid allows a cusped bubble shape whereas a Newtonian fluid does not.

Experimental

This study focuses on a particular complex fluid, the micellar system CTAB/NaSal (Shikata et al. 1987). This is one of several aqueous solutions involving the organic salt sodium salicylate (NaSal), which facilitates the formation of long tubular “worm-like” micelles of cationic surfactants such as CTAB (Cates and Candau 1990). In these solutions, the long micelles fluctuate in length, and shear causes the formation of structures (Liu and Pine 1996; Boltenhagen et al. 1997) which are much larger than individual micellar tubes (Shikata et al. 1987; Keller et al. 1998). The CTAB and NaSal used here are obtained from Aldrich, and dissolved in distilled deionized water without further purification. The fluids are mixed for several days, then allowed to sit for a day before use. We restrict our solutions to the molar ratio 1:1 (Shikata et al. 1988), and have found that only for a range of concentrations around 10 mmol/l can we obtain both a low Reynolds number flow and a reasonably liquid mixture. The surface tension is measured with a du Nouy ring tensiometer (Fisher), and for $[\text{CTAB}] = [\text{NaSal}] = 10.7 \text{ mmol/l}$ we find that $\sigma = 35.8 \pm 0.1 \text{ dynes/cm}$ (averaged over three measurements). For the same solution, the density is $\rho = 1.05 \text{ g/cm}^3$.

In the standard experiments in which cusped bubbles have been seen, the bubble rises through a quiescent fluid. To allow for the detailed measurement of the bubble shape and cusp over long periods of time, we have developed a vertically rotating toroid in which the bubble remains stationary in the lab frame. A schematic diagram is shown in Fig. 1a. Two PVC cylinders are held concentrically between clear polycarbonate endplates, and the cell cavity has an outer radius $R = 10.0 \text{ cm}$, radial gap $\Delta R = 4.3 \text{ cm}$, and axial width of 4.2 cm, with a mean circumference of about 50 cm. A

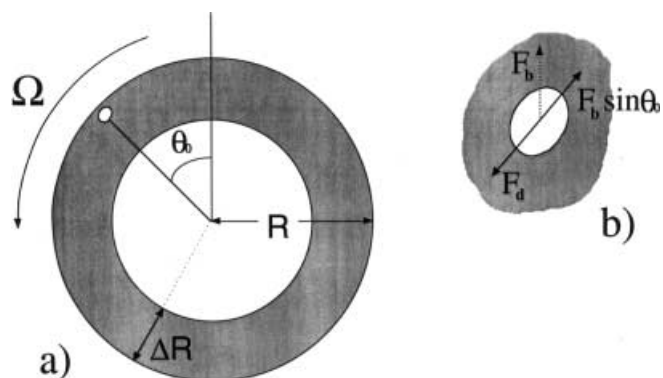


Fig. 1 **a** Diagram of the vertically rotating toroidal cell: the bubble sits at angle θ_0 , while the cell rotates with frequency Ω . **b** Buoyant and drag force components acting on the bubble

small air bubble of known volume is created in the fluid using a syringe. The cell is then rotated vertically at a fixed frequency Ω by a precision brushless servo drive through a 60:1 or 120:1 gearbox. The rotating cylinders entrain the interior fluid to undergo approximately rigid body rotation. The bubble is displaced downward in the cell by the drag of the rotating fluid until the azimuthal component of the bubble's buoyancy F_b matches the drag F_d at some angle θ_0 (see Fig. 1b). When Ω is such that $\theta_0 = 90^\circ$, the entire buoyancy of the bubble is balanced by the drag, as in free rise, and the bubble is radially centered in the gap between the cylinders; this is the Ω at which we work. The equivalent terminal velocity of the bubble is then given by $U = 2\pi(R - \Delta R/2)\Omega$. Note that we consider only the asymptotic state of the bubble motion, and ignore any spin-up effects (Shankar Subramanian 1992). If the rotation rate Ω is too high, the bubble is entrained and rotates more or less with the fluid. The bubble image is recorded with a CCD camera to videotape or directly to a computer via a frame grabbing card. The experimental measurements are obtained from video images of the interface shape.

The trade-off for the bubble's stationarity in the lab frame is that it is moving in a curved geometry, which may affect its exact shape. However, the bubble diameter is usually much smaller than the cell cross section. These complicating factors are being analyzed using the exact equations for our experiment to derive corrections due to the cell curvature, and experimentally by studying wall effects.

We have also run conventional experiments on rising bubbles to compare with the rotating cell. The stationary cell is an acrylic plastic box of height 40 cm and cross-sectional area $5 \text{ cm} \times 13 \text{ cm}$. Air is injected at the bottom through a needle by a calibrated syringe. This cell is used for qualitative observations only, confirming that all of our observations, especially the bubble oscillations, also occur for an actual rising bubble.

Results

As a test of our experimental technique, we first confirm that the standard cusp transition for rising bubbles also occurs for a bubble held stationary in our rotating cell. We have measured air bubbles in a standard weakly elastic power law fluid (Chhabra 1993; Acharya et al. 1977), an 0.8% solution of carboxymethylcellulose (CMC) in a 50:50 water/glycerol mixture. For each volume, the equivalent terminal bubble velocity U is shown in Fig. 2a. An example of a bubble in our experiment is shown in Fig. 2b. A discontinuity in the velocity-volume relation of magnitude $\Delta U \simeq 4 \text{ cm/s}$ occurs at the same volume at which the cusp appears ($V_b \simeq 0.2 \text{ cm}^3$). Both of these values are of the same size as seen in other experiments on rising bubbles (Astarita and Apuzzo 1965; Liu et al. 1995; Leal et al. 1971), in particular for CMC solutions (Acharya et al. 1977).

In the aqueous micellar system CTAB/NaSal, we have observed cusped bubbles for two different concentrations: $[\text{CTAB}] = [\text{NaSal}] = 8.0 \text{ mmol/l}$ and 10.7 mmol/l . Surprisingly, for both solutions we find that some bubbles undergo relaxation-type oscillations during which a cusp is drawn out and then rapidly snaps back. A sequence of images illustrating such an oscillation is shown in Fig. 3, representing only about 50% of the total oscillation period of 1.24 s. There is an apparent critical volume, $V_0 \simeq 0.03 \text{ cm}^3$ for $[\text{CTAB}] = 10.7 \text{ mmol/l}$, below which neither cusp nor oscillations

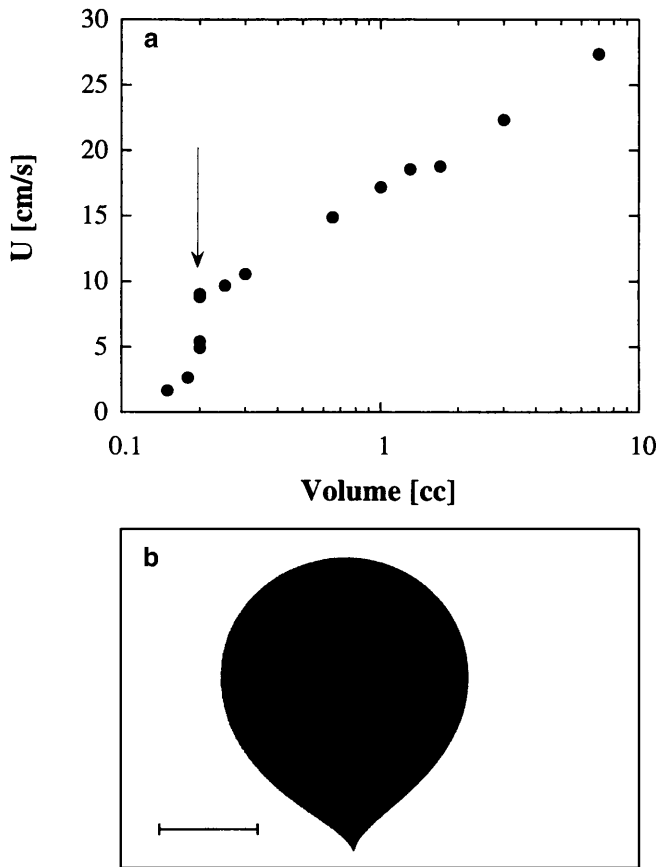


Fig. 2a, b Experiments on stationary “rising” bubbles in rotating cell with 0.8% CMC in 50:50 water/glycerol: **a** the fluid speed when $\theta_0 = 90^\circ$, equivalent to the bubble rising speed, vs the bubble volume (the *arrow* indicates the transition from rounded to cusped bubbles); **b** image of a bubble with $V_b = 0.30 \text{ cm}^3$ (the *scale* shown is 3 mm across)

are observed; just at onset the oscillations are rare and very irregular in timing.

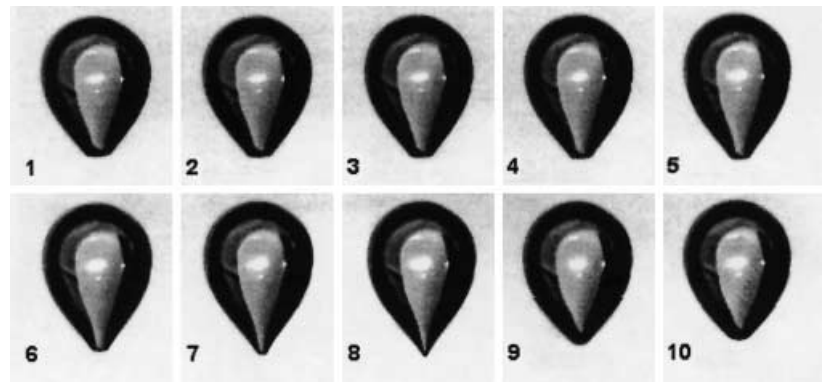
To compare the buoyancy of the bubble with the surface tension forces which apparently cause the cusp to snap back, we evaluate the Bond number $Bo \equiv$

$\rho g r_e^2 / \sigma$, where $r_e = (3V_b/4\pi)^{1/3}$ is the effective radius of the bubble. At the onset of the oscillations ($r_e \simeq 0.19 \text{ cm}$), we find that $Bo \simeq 1.1$ for $[\text{CTAB}] = 10.7 \text{ mmol/l}$. The criterion $Bo \sim 1$ has often been taken as an estimate of the Hadamard-Rybczynski transition for rising bubbles (Chhabra 1993; Acharya et al. 1977; Bond and Newton 1928), though it has been argued that this criterion is inappropriate for the occurrence of the velocity discontinuity (and cusp onset) (Liu et al. 1995), as it does not include any reference to non-Newtonian effects (Rodrigue and De Kee 1999; Rodrigue et al. 1996). It may thus be fortuitous that we find $Bo \simeq 1$ at this transition, and we are checking this result at other concentrations. We estimate the Reynolds number Re of the flow by assuming that the buoyancy of the rising bubble is balanced by a drag force $4\pi\mu_e U r_e$ (Chhabra 1993), where μ_e is the effective viscosity defined by this relation. We find $Re \leq 5 \times 10^{-2}$ for the results reported here.

In the course of an oscillation the bubble tail appears to assume a flattened knife-edge shape (Fig. 3, frames 1–7), as seen by Hassager (1979) and studied extensively by Liu et al. (1995). However, at its greatest extension, the cusp appears pointlike (frame 8). We are currently investigating the three-dimensional structure of the oscillating shape.

To quantify these oscillations we measure the streamwise length of the bubble as a function of time, as shown in Fig. 4. The asymmetry of these oscillations represents the rapid snap back from the cusp shape (Fig. 3, frames 8–9), which occurs in less than 1/30 of a second (a single video frame). Although the oscillations are never perfectly periodic, we find that the average frequency f_A is a well-defined function of volume. The oscillations are faster for larger bubbles, which have a larger average velocity. Figure 5 shows a plot of f_A vs the difference between the bubble volume and its onset value, $V_b - V_0$. This dependence is not unambiguously resolved in our experiment, although there is some indication of scaling. We find that $f_A \sim (V_b - V_0)^\alpha$, with $\alpha = 0.38 \pm 0.07$ over the range of our data.

Fig. 3 Sequential images of a bubble in the rotating cell. The images shown are every 67 msec. ($V_b = 0.15 \text{ cm}^3$, 10.7 mmol/l CTAB/NaSal solution)



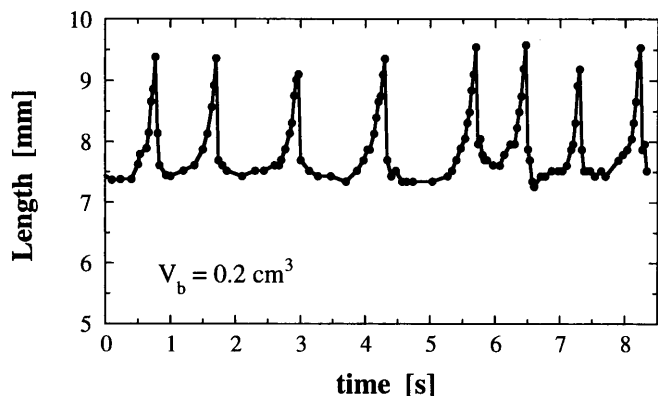


Fig. 4 Streamwise length oscillations of an 0.2 cm^3 bubble (10.7 mmol/l CTAB/NaSal)

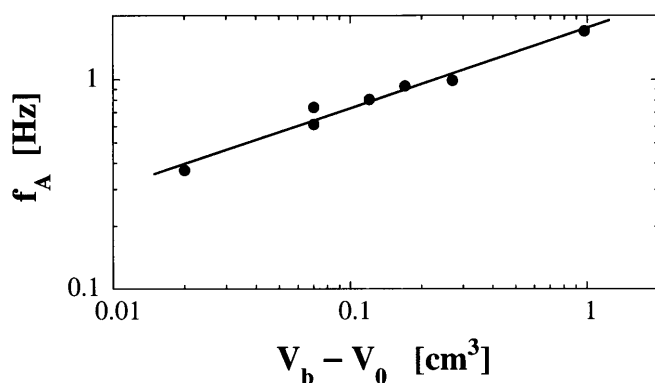
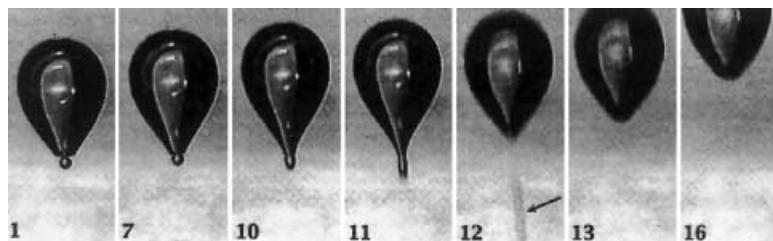


Fig. 5 Average oscillation frequency f_A vs volume difference $V_b - V_0$, with $V_0 = 0.03 \text{ cm}^3$. The line shown corresponds to the fitted scaling $f_A \sim (V_b - V_0)^\alpha$, with $\alpha = 0.38$

The self-oscillations in the shape of the bubble are also coupled to its motion, which we observe in both experimental cells. As the bubble length increases, and the cusp begins to form, the velocity decreases. When the length decreases suddenly (as the cusp snaps back), the bubble accelerates and jumps upwards. Then it again slows down, and the length begins to grow as the cycle repeats. During the jump a recoil is noticeable in the fluid behind the bubble, an indication of a negative wake

Fig. 6 Sequential images of the repulsion of a smaller bubble in the wake of a large bubble which has jumped, evidence of an unsteady negative wake. The images are labeled in units of $\Delta t = 33 \text{ msec}$, and the arrow in frame 12 indicates the blurred image of the ejected smaller bubble (10.7 mmol/l CTAB/NaSal solution; larger bubble $V_b = 0.05 \text{ cm}^3$)



similar to that seen for cusped bubbles in conventional polymer fluids (Hassager 1979), though here the wake is oscillating. If a second smaller bubble happens to be present, this recoil can result in its sudden downward repulsion, a remarkable example of which is shown in Fig. 6.

Discussion

We do not at present have a consistent explanation for the observed oscillations of bubbles rising in a micellar solution. However, we can identify two possible directions from which an explanation might come, as discussed below.

In the flow of non-Newtonian or viscoelastic fluids under steady conditions, the appearance of macroscopic oscillations, such as the sharkskin and melt fracture instabilities seen during extrusion, is a striking manifestation of the underlying complex molecular structure (Bird et al. 1987; Denn 1990). The surface oscillations of sharkskin are often modeled by a non-monotonic relation between an averaged shear stress and shear rate (see for example Larson 1988; Malkus et al. 1991; Renardy 1995; Yang et al. 1988), which includes a transition from no-slip to slip at the extruder wall (Denn 1990). Similar oscillations are seen in the Johnson-Segalman-Oldroyd model, which includes an intrinsic non-monotonic relation between stress and shear rate (Malkus et al. 1991). The self-assembling wormlike micellar solutions such as those studied here are also thought to have a nonmonotonic stress/shear rate relation (Spenny et al. 1993; Porte et al. 1997), which may be the cause of the oscillations we observe. The relevant aspect is the existence of a range of shear rates which are not accessible in the fluid, where the stress is a decreasing function of shear rate (Porte et al. 1997; Cappelaere et al. 1997). The rising bubble's periodic distortion could be the result of a jump across this region, overshooting a balance condition with the surface tension which would normally lead to a steady cusp-like tip. The rapid snap-back of the cusp would then be surface tension driven, an aspect which could be tested.

It is also possible that these oscillations are related to the formation of mesoscale inhomogeneities in these micellar systems, the so-called “shear-induced structures” (Rehage and Hoffmann 1982; Cates and Candau 1990). There is increasing experimental evidence of the onset of periodic oscillations in the flow of wormlike micellar solutions under steady conditions, including observations of periodic growth and tearing of small structures (Liu and Pine 1996) and time dependent flow (Kock et al. 1998) in a Couette cell, and oscillating rings in a constant stress parallel plate rheometer (Wheeler et al. 1998). It is also known that there is a shear-induced transition from an isotropic to a nematic ordering of these micelles (Berret et al. 1994; Cappelaere et al. 1997). The rising bubble may be creating such structures in its wake from which it periodically breaks free, causing the oscillations. We are currently investigating this suggestion experimentally.

Conclusion

We have performed experiments on the motion and shape of rising gas bubbles in non-Newtonian fluids by employing a novel geometry in which the interface remains fixed in the lab frame. In this apparatus, the well-known transition to a cusped bubble and the accompanying discontinuity in the velocity as a function of bubble volume are observed in a CMC solution. By studying the wormlike micellar solution CTAB/NaSal, we have observed cusped bubbles which oscillate in shape as they rise. The frequency of these oscillations increases with bubble volume.

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