



Influence of surfactants on the electrohydrodynamic stretching of water drops in oil



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ARTICLE INFO

Article history:

Received 20 January 2017

Revised 30 June 2017

Accepted 25 August 2017

Available online 1 September 2017

Keywords:

Electrohydrodynamics

Droplet

Surfactants,

ABSTRACT

In this paper we present experimental and numerical studies of the electrohydrodynamic stretching of a sub-millimetre-sized salt water drop, immersed in oil with added non-ionic surfactant, and subjected to a suddenly applied electric field of magnitude approaching 1 kV/mm. By varying the drop size, electric field strength and surfactant concentration we cover the whole range of electric capillary numbers (Ca_E) from 0 up to the limit of drop disintegration. The results are compared with the analytical result by Taylor (1964) which predicts the asymptotic deformation as a function of Ca_E . We find that the addition of surfactant damps the transient oscillations and that the drops may be stretched slightly beyond the stability limit found by Taylor. We proceed to study the damping of the oscillations, and show that increasing the surfactant concentration has a dual effect of first increasing the damping at low concentrations, and then increasing the asymptotic deformation at higher concentrations. We explain this by comparing the Marangoni forces and the interfacial tension as the drops deform. Finally, we have observed in the experiments a significant hysteresis effect when drops in oil with large concentration of surfactant are subjected to repeated deformations with increasing electric field strengths. This effect is not attributable to the flow nor the interfacial surfactant transport.

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1. Introduction

Surfactants are ubiquitous in two-phase fluid flows. Take for instance a single drop falling through a viscous fluid, perhaps the simplest and most widely studied two-phase flow configuration. While the classic results by Hadamard (1911) and Rybzyński (1911) give the analytical result for the flow field in this case, experimental investigations mostly fail to agree with this result. The discrepancy is attributed to trace surface-active contaminants, found even in the most purified of liquids. It is natural, then, also to consider the effects of surfactants on the more complicated case of electrohydrodynamic deformation of a conducting drop falling in an insulating oil.

The case of a drop deforming in an electric field is interesting, not only as an intriguing physical phenomenon of which our understanding can be improved, but also for applications e.g. to chemical processing equipment such as electrocoalescers (Atten, 1993; Eow et al., 2001; Lundgaard et al., 2006). A deeper understanding

of the physical processes at play in this system could lead to improved coalescer equipment and reduced emissions.

We will consider here experiments and simulations of sub-millimetre-sized drops of brine falling in a highly refined oil with added surfactant, studying the drop deformations and oscillations induced by square voltage pulses of varying amplitude applied to parallel electrodes above and below such a drop.

When performing these studies of drop deformations, it is crucial to have a system which is well characterised in terms of the fluid and the interfacial properties. To overcome the uncertainties associated with unknown trace contaminants acting as surface-active agents, we deliberately add a non-ionic surfactant (Span 80) in known, small quantities. The interfacial tension as a function of surfactant concentration is then measured, together with the bulk properties, to give a well-characterised system.

There is a large amount of research on the deformations of drops in electric fields, using analytical, experimental and numerical techniques; we will not summarise all of it here. The review by Melcher and Taylor (1969) covers the fundamentals in a thorough fashion, while the review by Saville (1997) gives an update with more recent results in the field. However, when surfactants

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are added to this picture, the literature is not so extensive. Previous authors (Ha and Yang, 1998; Zhang et al., 2015) have investigated the influence of surfactants on the electrohydrodynamic stretching experimentally, but they have been limited to considerations of the static (equilibrium) deformation, as well as drop sizes above 1 mm in diameter, and a limited number of observations. Computational studies in the literature, namely previous work by Teigen et al. (2010), and the paper by Nganguia et al. (2013) which finds good agreement with Teigen et al. (2010), have also been focused on the static deformation. Note that the numerical code used in this paper is the same as in Teigen et al. (2010).

Taking a step further, we consider here also the dynamical behaviour of the stretching drops, in particular the effects of the surfactant concentration on the damping of the drop oscillations. We work with drops smaller than 1 mm in diameter. We report results for many drop deformations, almost 300 for the experiments and 44 representative cases for the simulations.

This work is an extension of our initial investigation (Ervik et al., 2014), where five cases of the electrohydrodynamic deformation of drops in insulating oil were studied. In the present work we have extended this analysis to a parameter study of the factors influencing the deformation and the deviations from the classical result by Taylor (1964), which does not take surfactants into account. The analytical result by Taylor has been found to agree very well with subsequent results, see e.g. Brazier-Smith (1971), and for this reason we use it as a supporting line in the plots and analysis throughout the paper. Following Taylor, we use the dimensionless electric field strength $\zeta = \sqrt{Ca_E}$ in the following.

The results presented here show that the deviation from Taylor's expression is negligible below dimensionless electric field strengths of $\zeta \approx 0.4$, while above this threshold they become significant. We demonstrate that drops in the presence of surfactants may be deformed beyond the stability limit given by the Taylor theory. Finally we study the effect of the surfactant concentration, and the effects of Marangoni stresses on the damping of drop oscillations. Our results indicate that small concentrations of surfactant give a significant increase in the damping whilst having but a small effect on the equilibrium (static) shape. Also, for the highest surfactant concentration used here, we observe in the experiments a significant hysteresis effect of repeated stretchings. This effect is not seen in the simulations, so it cannot be explained by the hydrodynamics and the surfactant transport processes which are modelled by our approach.

2. Theory

The flow of single-phase oil or water can be described by the incompressible Navier–Stokes equations

$$\nabla \cdot \mathbf{u} = 0, \tag{1}$$

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\frac{\nabla p}{\rho} + \frac{\eta}{\rho} \nabla^2 \mathbf{u} + \mathbf{f}, \tag{2}$$

where \mathbf{u} is the velocity field, p is the pressure, ρ is the density, η is the dynamic viscosity, and \mathbf{f} is the acceleration caused by some body force, e.g. the gravitational acceleration. This description can be extended to a two-phase flow by incorporating three things, namely that there is an interface separating the two fluids, that the fluids may have different viscosities η_1, η_2 and densities ρ_1, ρ_2 , and finally the effects of interfacial tension and interfacial tension gradients. We mark the drop properties with subscript $_1$ and the bulk properties with $_2$, and denote the interfacial tension by γ . The viscosity difference and the interfacial tension γ contribute to jumps across the interface in various properties such as the pressure; this is detailed in Eqs. (16)–(18) below. Mathematically, this

can be incorporated into the Navier–Stokes equations as a singular contribution to \mathbf{f} in Eq. (2).

This system admits two dimensionless groups, which we may take to be the Reynolds number Re and the Ohnesorge number Oh . The Reynolds number is of interest for a falling drop, where it is defined as $Re_D = \rho_2 u_T D / \eta_2$, u_T being the terminal velocity and D being the drop diameter. For the drops considered here, the Reynolds number is small ($Re_D < 1$), meaning that the inertial term in Eq. (2) is unimportant for the flow at terminal velocity.

For an oscillating drop, the Ohnesorge number is an important quantity; some authors use the inverse of the Ohnesorge number as the “oscillation Reynolds number” Re_{osc} . We use the definition $Oh = \eta_2 / \sqrt{\rho_2 \gamma D}$, since the ambient fluid is much more viscous for the cases considered here. For the oscillations, the Ohnesorge number is also small ($Oh < 0.2$), but here the inertial term is important since small Oh corresponds to large Re_{osc} .

When considering a single small (i.e. spherical) drop falling in a clean fluid at low Reynolds number, the terminal velocity as well as the flow in the entire domain is given analytically by the results that Hadamard (1911) and Rybzyński (1911) obtained independently,

$$v_{T,HR} = \frac{(\rho_1 - \rho_2)gD^2(\eta_1 + \eta_2)}{6\eta_2(3\eta_1 + 2\eta_2)}. \tag{3}$$

Experimental results for the terminal velocity, however, tend to not agree with this result (see e.g. Bond and Newton, 1928, Fig. 1), but a closer agreement is found with the formula derived by Stokes (1851) for a hard sphere falling in an unbounded domain,

$$v_{T,S} = \frac{(\rho_1 - \rho_2)gD^2}{18\eta_2}. \tag{4}$$

Indeed Hadamard himself was aware of this discrepancy, as he states in his 1911 paper.

We note that for $\eta_1 < \infty$, the graphs of $v_T(D)$ given by Eqs. (3) and (4) only intersect at $D = 0$, and thus the terminal velocity of a falling drop is an observable quantity that can determine if a system is clean or not. An experimental observation closer to Eq. (4) indicates a contaminated system, which is indeed the observation for most fluid combinations. It is noteworthy that the experiments which have obtained values agreeing with Eq. (3) are for quite singular fluid combinations, e.g. mercury drops in glycerine (Levich, 1962).

The currently accepted explanation (see Clift et al., 1978, pp. 35–41) of this phenomenon is that trace contaminants in the system act as surfactants which are swept along the interface by the flow, creating an interfacial-tension gradient which results in a Marangoni force, with the end result that the drop interface is immobile. Since the nature of these trace contaminants are not known, we deliberately add to the oil a known amount of a non-ionic surfactant, Span 80, such that we obtain a well-described fluid system.

The interfacial tension, γ , can be related to the bulk concentration of surfactant, Λ , using the Szyszkowski (1908) equation of state (EoS):

$$\gamma(\Lambda) = \gamma_0 \left[1 - \beta \ln \left(1 + \frac{\Lambda}{a_L} \right) \right], \tag{5}$$

where γ_0 is the interfacial tension without surfactants, $\beta = R_{gas} T \Gamma_\infty / \gamma_0$ is the interfacial elasticity, and $a_L = k_{des} / k_{ads}$ is the ratio between the adsorption and desorption coefficients of the surfactant. In the expression for β , Γ_∞ is the maximum possible interfacial concentration of surfactant, R_{gas} is the universal gas constant, and T is the temperature (in Kelvin). The parameters β , a_L of this EoS may be computed by fitting to experimental data; note that this also determines Γ_∞ when the temperature is known.

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