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Effect of surfactant on the break-up of a liquid thread: Modeling, computations and experiments

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ABSTRACT

The effect of insoluble surfactant on the breakup of an inviscid jet surrounded by a viscous fluid at low Reynolds number is studied both theoretically and experimentally. Equations governing the evolution of the interface and surfactant concentration are derived using long wavelength approximations. Numerical simulations of this long-wave asymptotic model show that the presence of surfactant at the interface retards the pinch-off and a slender quasi-steady thread is formed. After the formation of the thread, the diffusion of surfactant plays an important role and causes the jet to pinch. Experiments are performed and the results are presented which show that the presence of surfactant slows down the breakup of the bubble. We used high speed digital imaging to capture details of the pinching bubble in a glycerol/water mixture in the absence or presence of surfactants. Digitized video images of bubble elongation are captured sequentially in time and analyzed using image analysis software. It is found that the presence of surfactant slows down the breakup of a jet in a highly viscous exterior fluid. Experimental findings are in agreement with the theoretical and computational results.

1. Introduction

The problem of jet breakup has been extensively studied both experimentally and theoretically for jets injected into air, where the air is modeled as a vacuum. For the case of these single fluid jets, theoretical developments started with the classical work of Rayleigh [1] who studied the breakup of an infinite inviscid jet injected into air linear stability theory. Weber [2] extended the linear theory of Rayleigh to the case of viscous jets. In both of the above studies, it was shown that a cylindrical fluid column is unstable under axisymmetric perturbations whose wavelength is greater than the circumference of the undisturbed jet. After the pioneering work of Rayleigh and Weber, subsequent numerical and theoretical investigations [3,4] have focused on elucidating details of the breakup of single fluid jets. Eggers [5] presented a comprehensive review of non-linear theories and experimental studies on the surface tension driven single fluid jet, including pinch-off.

For the case of two fluid jets, the effects of the surrounding fluid must be included in the analysis of the jet instability and breakup. Lister and Stone [6] considered the effect of a viscous external fluid on the non-linear dynamics and breakup of a fluid jet evolving under Stokes flow. They found that when the external fluid viscosity is introduced, the dynamics near the pinch-off is given by a balance between capillary pressure, interior and exterior viscous stresses. Doshi et al. [7] used experiments, large scale numerical simulations and asymptotic analysis to study the breakup of a low viscosity jet inside a highly viscous exterior fluid and showed that different initial and boundary conditions lead to different asymptotic profiles when the interior viscosity is small enough.

The presence of surfactants at the interface can significantly alter the interfacial properties. There are some studies to understand the influence of surfactants on the breakup of two fluid jets. Timmermans and Lister [8] also used linear stability analysis as well as one-dimensional approximations to Navier–Stokes equations to study the surface tension driven flow of a liquid jet in inviscid surroundings. They concluded that for a fluid jet in inviscid surroundings, surfactants have little effect on the pinch-off since the flow in the jet sweeps the surfactant away from the pinch point due to advection. However, this is not necessarily the case when there is no internal fluid, or the internal fluid is much less viscous than the outer fluid as shown in the present work. Stone et al. [9] considered the effect of surfactant on the deformation and breakup of a drop in extensional flow and established a qualitative relationship between the flow, surfactant concentration

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and surface stresses. Wang [10] investigated the effect of surfactant on the moving contact line on inclined plane.

In this work, we use long-wave asymptotic analysis and experiments to study the effect of surfactants on the breakup of a two-fluid jet. We consider an inviscid fluid jet in a highly viscous exterior fluid. To find the solution of the Stokes equations, we use a novel approach based on the slender body theory, which exploits the large aspect ratio of the jet. Due to linearity of the Stokes equations, an exact solution can be written as an integral of a distribution of point forces, or Stokeslets over the boundary of a fluid region. This approach introduces an integral equation with a modified Stokeslet kernel on the filament centerline that relates the filament forces to the velocity of the centerline. Slender body theory has been successfully employed to study Stokes flow in different geometries [11–13].

We also performed experiments, which support the predictions of the model. On the experimental front, we used high speed digital camera to capture the details of the pinching jet. Air bubbles are created in a glycerol/water mixture in the absence or presence of surfactants. Digitized video images of the bubble elongation and breakup were captured sequentially in time and analyzed using image analysis software. It is found that the presence of surfactant slows down the pinch-off in viscous exterior fluid.

2. Mathematical model

We consider an infinite, axisymmetric, periodic fluid jet surrounded by a viscous fluid. The jet is assumed to be slender, i.e., the wavelength of the jet is much larger than its average width. We consider the interior of the jet to be inviscid, we can think of it as an analogue of a long air bubble in viscous surrounding. The jet is assumed to be axisymmetric with respect to the axial coordinate axis, which corresponds to the axis of symmetry. The surrounding fluid is assumed to be viscous, incompressible and Newtonian. Considering a cylindrical coordinate system (r, θ, z), the surface of the jet is denoted by $r = \epsilon R(z,t)$ and the axial and radial components of velocity are denoted by u_z and u_r respectively. Neglecting inertia, the non-dimensional governing equations are axisymmetric Stokes equations and the continuity equation given as

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u_z}{\partial r}\right) + \frac{\partial^2 u_z}{\partial z^2} = \frac{\partial p_o}{\partial z}, \tag{1}$$

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial u_r}{\partial r}\right) - \frac{u_r}{r^2} + \frac{\partial^2 u_r}{\partial z^2} = \frac{\partial p_o}{\partial r}. \tag{2}$$

$$\frac{\partial u_z}{\partial z} + \frac{1}{r}\frac{\partial}{\partial r}(r u_r) = 0. \tag{3}$$

The non-dimensional tangential stress balance, normal stress balance and kinematic condition at the interface $r = \epsilon R(z,t)$ are given by

$$\frac{2}{(1+\epsilon^2 R'^2)}\left[\epsilon R'\frac{\partial u_r}{\partial r} + (1-\epsilon^2 R'^2)\frac{1}{2}\left(\frac{\partial u_r}{\partial z} + \frac{\partial u_z}{\partial r}\right) - \epsilon R'\frac{\partial u_z}{\partial z}\right] = -\frac{\partial \sigma}{\partial z}\frac{1}{\sqrt{1+\epsilon^2 R'^2}}, \tag{4}$$

$$p_i - \epsilon p_o + \frac{2\epsilon}{(1+\epsilon^2 R'^2)}\left[\frac{\partial u_r}{\partial r} - \epsilon R'\left(\frac{\partial u_r}{\partial z} + \frac{\partial u_z}{\partial r}\right) + \epsilon^2 R'^2\frac{\partial u_z}{\partial z}\right] = \frac{\sigma}{R\sqrt{1+\epsilon^2 R'^2}}\left[1 - \frac{\epsilon^2 R R''}{1+\epsilon^2 R'^2}\right], \tag{5}$$

$$u_r = R_t + \epsilon R' u_z. \tag{6}$$

Here σ is represents variable surface tension and is a decreasing function of surfactant concentration, ϵ represents the aspect ratio of a slender jet, p_i and p_o are inside and outside pressure respectively. We will base our asymptotic analysis on this slenderness parameter. The relationship between the surface tension and the surfactant concentration is given by the equation of state $\sigma = \sigma(\Gamma)$. These are determined empirically. There are several different kinds of linear and non-linear relationships which are used. In this study, we use a non-linear dependance of σ on Γ, also known as the Langmuir equation, given in non-dimensional form as

$$\sigma = 1 + \beta \ln(1-\Gamma), \tag{7}$$

where $\beta = \frac{R_g T \Gamma_\infty}{\sigma_0}$, it represents the sensitivity of the surface tension to the surfactant concentration, σ_0 is the surface tension of the clean interface, Γ_∞ is the maximum allowable interfacial concentration of surfactant (the maximum packing density), R_g is the universal gas constant and T represents the absolute temperature.

In order to quantify the effect of surfactant on the interface, we introduce a convection diffusion equation which governs the surfactant transport along the interface. The presence of the exterior fluid causes the surfactant to spread across the interface in a non-uniform way. There are other factors as well that might influence the surfactant distribution such as diffusion and the evolution of the interface (e.g., stretching or contracting). Wong, Rumschitzki and Maldarelli [14] suggested a convection diffusion equation which governs the surfactant transport along the interface for the evolution of the surfactant concentration for a general parametric representation of the interface $\mathbf{X}(\alpha, t)$.

$$\left.\frac{\partial \Gamma}{\partial t}\right|_\alpha - \left.\frac{\partial \mathbf{X}}{\partial t}\right|_\alpha \cdot \nabla_s \Gamma + \nabla_s \cdot (\Gamma \mathbf{u}_s) - D_s \nabla_s^2 \Gamma + \Gamma \kappa \mathbf{u} \cdot \mathbf{n} = 0, \tag{8}$$

where Γ is the surfactant concentration, $\left.\frac{\partial}{\partial t}\right|_s$ denotes the time derivative keeping the arc length coordinate fixed, $\nabla_s = (\mathbf{I} - \mathbf{nn}) \cdot \nabla$ is the surface gradient operator, $\mathbf{u}_s$ is the velocity at the interface, κ is the mean curvature of the interface and D_s is the interfacial surfactant diffusivity. In this study, we have considered the surfactant to be insoluble, i.e., there is no net flux of the surfactant to and from the interface to the bulk fluid. The non-dimensional surfactant transport equation in cylindrical coordinates is given as

$$\frac{\partial \Gamma}{\partial t} + \frac{\epsilon}{R\sqrt{1+\epsilon^2 R'^2}}\frac{\partial}{\partial z}\left[\frac{R\Gamma}{\sqrt{1+\epsilon^2 R'^2}}(\epsilon u_r R' + u_z)\right] - \frac{\epsilon^2 R'}{1+\epsilon^2 R'^2}\frac{\partial R}{\partial t}\frac{\partial \Gamma}{\partial z} - \frac{\epsilon}{Pe_s}\left[\frac{1}{R\sqrt{1+\epsilon^2 R'^2}}\frac{\partial}{\partial z}\left(\frac{R}{\sqrt{1+\epsilon^2 R'^2}}\frac{\partial \Gamma}{\partial z}\right)\right] + \frac{\Gamma(u_r - \epsilon R' u_z)}{R(1+\epsilon^2 R'^2)}\left[1 - \frac{\epsilon^2 R R''}{1+\epsilon^2 R'^2}\right] = 0. \tag{9}$$

where we have introduced a non-dimensional surface Peclet number $Pe_s = \frac{\sigma_o L}{D_s \mu}$. The interior fluid is inviscid, incompressible and the total volume of the bubble is conserved. We use this as a accuracy check for our numerical computations. A second conserved quantity in the system is the total amount of surfactant, which satisfies the relation

$$\int_S \Gamma dS = 4\pi\chi,$$

where $\chi = \frac{\Gamma_i}{\Gamma_\infty}$, and Γ_i is the average over the bubble surface of the initial surfactant concentration, Γ_∞ represents the upper bound to the surfactant concentration [9].