ABSTRACT

Wee, Hansol Ph.D., Purdue University, May, 2023. Effects of surface rheology in free surface flows. Major Professor: Osman A. Basaran

Adsorption onto and lowering the surface tension of a fluid interface by surfactants are exploited in applications, such as enhanced oil recovery, coating flows, lung surfactants, and drop/jet breakup. However, surfactant concentration can be nonuniform at the interface because surfactant molecules can be transported along it by convection and diffusion and also due to normal dilatation and tangential stretching of the interface. Thus, aside from simply lowering surface tension, nonuniformity in surfactant concentration causes gradients in surface tension and gives rise to tangential interfacial (Marangoni) stresses. The latter brings about rich physics including tears of wine, interfacial turbulence in mass transfer, and droplet bouncing. In addition to lowering surface tension and the Marangoni effect, surfactants may also induce surface rheological effects as surfactant molecules deform against each other. In this thesis, surface rheological effects in free surface flows are examined through both analytical and numerical treatment of the incompressible Navier-Stokes equations subjected to the traction boundary condition augmented by the Boussinesq-Scriven equation to account for surface viscous effects. Rigorous and robust numerical algorithms based on Galerkin finite element (GFEM) method are developed for predictions of surfactant transport, surface rheological effects and hydrodynamics in response to the motion of moving boundaries. The accuracy of computational predictions is verified by demonstrating that computed results accord well with scaling theories.

The breakup of surfactant-covered liquid jets or threads is examined in the presence of surface rheological effects. During the thinning of the thread, convection and diffusion—the relative importance of which is characterized by a Peclet number Pe—compete to determine the distribution of surfactant along the interface. As fluid evacuates the thinning neck, surfactant is convected away from that locale. However, the resulting concentration gradient gives rise to diffusion which tries to replenish the neck with surfactant. First, the breakup of a Stokes thread covered with a monolayer of insoluble surfactant is examined when either surfactants are convected away from the space-time singularity ($Pe \gg 1$) or

diffusion is dominant ($Pe \ll 1$). Surprisingly, in both limits, surface rheological effects always enter the dominant balance of forces and alter the thread's thinning rate. When surface rheological effects are negligible, regardless of Pe, the dynamics in the vicinity of the finite-time singularity is self-similar and, asymptotically, there is always a transition from a diffusion-dominated scaling regime to a convection-dominated one as the space-time singularity is approached.Theory and simulations are used to show that a Stokes thread undergoing breakup under conditions in which surface viscous stresses are present gives rise to an exceptional type of dynamics in the vicinity of the neck. In contrast to previous studies of breakup in which there is always a dynamical transition between different scaling regimes as pinch-off nears, presence of surface viscous stresses results in dynamics that cuts off this universal type of response. It is demonstrated that when $Pe < Pe_c$, the dynamics is always diffusion-dominated and that the rate of thread thinning is exponential in time. If, however, $Pe > Pe_c$, the dynamics is self-similar and exhibits power-law dependence on time until pinch-off. While all of the aforementioned studies are concerned with the dynamics of a highly viscous liquid thread or jet, inertia can never be identically zero for real fluids. Indeed, it is now well-known that jet breakup when the interface is either clean or covered with surfactants but where surface rheological effects are absent must asymptotically always involve inertia. Thus, it is necessary to extend the results obtained from Stokes flow to situations in which inertia is present. A combined asymptotic and numerical analysis reveals that Marangoni stresses become negligible compared to other forces and the rate of jet thinning is significantly lowered by surface viscous effects.

Rupture of thin free films of Newtonian fluids is analyzed when the sheets' two free surfaces are covered with insoluble surfactant and surface rheological effects are important. The analysis relies on a long-wavelength model comprised of a system of one-dimensional evolution equations for film thickness $h(z,t)$, lateral velocity $v(z,t)$ and surfactant concentration $\Gamma(z,t)$ (z: lateral coordinate, t: time). For both highly viscous fluids in the Stokes limit and moderately viscous fluids, it is shown that the dominant balance involves van der Waals pressure and bulk viscous as well as surface viscous stresses while surface tension pressure and Marangoni stress are negligible.

Lastly, in a growing number of applications involving breakup of drops and jets of liquid metals, the working fluids have viscosities comparable to but surface tensions and densities much larger than water. The dynamics of thinning and pinch-off is governed by the Ohnesorge number Oh (dimensionless viscosity). When $Oh \ll 1$, the thread initially thins as if it were inviscid and its minimum radius obeys a universal scaling law $\tilde{h}_{min} = A\,(\gamma/\rho)^{1/3}\,(\tilde{t}_b - \tilde{t})^{2/3}$ where $\tilde{t}_b$ is the time $\tilde{t}$ at which the thread breaks up and $A \doteq 0.717$. Using high-accuracy simulation based on a sharp-interface algorithm, we show that, for sufficiently small Oh, the value of A predicted from computations agrees with the theoretical value to three decimal places and the inviscid power-law behavior can be observed over 2-3 decades in $\tilde{h}_{min}$ as $\tilde{t}_b - \tilde{t} \to 0$.