J. Non-Newtonian Fluid Mech. 136 (2006) 17–37

Journal of Non-Newtonian Fluid Mechanics

www.elsevier.com/locate/jnnfm

Dynamics of formation and dripping of drops of deformation-rate-thinning and -thickening liquids from capillary tubes

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Received 30 March 2004; received in revised form 30 January 2006; accepted 28 February 2006

Abstract

Dynamics of formation of drops of non-Newtonian liquids from capillary tubes is studied computationally. The rheology of the drop liquids is described by a constitutive relation that accounts for both deformation-rate-thinning and -thickening. The analysis is expedited by reducing the original system of three-dimensional but axisymmetric equations to a system of one-dimensional slender-jet equations. The slender-jet equations are solved by a method of lines using a finite element method for spatial discretization and an adaptive finite difference method for time integration. The simulations follow the formation in time of thousands of drops in sequence, including any satellites that may be produced upon the breakup of a thin thread connecting an about-to-form primary drop to the rest of the liquid attached to the tube. Rate-thickening is shown to produce bead-on-string patterns, which are typically attributed to viscoelastic effects, along the thin threads as they near pinch-off. Rate-thinning, on the other hand, is demonstrated to reduce the length of such thin threads. Simulations are used to identify conditions that may lead to minimization and/or elimination of unwanted satellites. Analysis of dripping or leaky faucets of non-Newtonian liquids reveals rich nonlinear dynamical behavior. As with Newtonian liquids, simple periodic or P-1, where P stands for period, dripping at low flow rates gives way to more complex responses as flow rate is increased. In addition to P-1, P-2, and P-4 responses seen in recent computational analyses of dripping faucets of Newtonian liquids, the new non-Newtonian simulations have also uncovered difficult-to-find P-3 responses as well as chaotic states. Rate-thinning and low viscosities are shown to enhance the complexity of observed responses. Rate-thickening, on the other hand, lowers the critical value of the flow rate for the onset of complexity but narrows the range of flow rates over which the dynamics is complex. The possibility of hysteresis is demonstrated and the effect of fluid rheology on the value of the flow rate for transition from dripping to jetting is determined.

Keywords: Drop formation; Dripping and leaky faucets; Ink jet printing; MEMS; Non-Newtonian fluids; Free surface flows; Nonlinear dynamics; Finite element analysis

1. Introduction

A wide variety of applications involve small-scale free-surface flows in which drops form from tubes [1]. Some examples include property measurement [2–4], combustion [5], atomization and spray coating [6,7], crop spraying [8], ink jet printing [9], printing of polymer transistors [10], and microarraying for genomics, combinatorial chemistry and drug discovery [11–13]. With rapid advances in drop-based technologies and their ever broadening scope of applications such as microarraying and printing of polymer circuits, there is renewed interest in the study of drop formation. The success of such cutting edge technologies depends strongly on the development of accurate methods of computing the dynamics of drop formation.

In many applications involving drops, and in particular ones used in printing and coating, the liquids encountered are non-Newtonian. For example, Pangalos et al. [14], who have carried out a detailed study of rheological properties of news inks of varying compositions, have found all of the inks that they studied to be rate-thinning. More recently, Fernandez et al. [15] have carried out a similar study and reported on rate-thinning as well as viscoelastic properties of highly pigmented inks. It is well-known that non-Newtonian liquids can exhibit responses that differ drastically from those of Newtonian liquids in most free surface flows and not just in drop formation. Rod climbing is but one example of such peculiar behavior (see, e.g., [16]). One of the most interesting effects encountered in stretching and/or thinning columns of non-Newtonian liquids is the formation of "bead-on-string" patterns [17–19]. Because these beads

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doi:10.1016/j.jnnfm.2006.02.009

can serve as precursors to undesirable satellites and spray, or mist, formation in applications, it is important to be able to predict their occurrences. Motivated by the widespread use of non-Newtonian liquids in drop formation applications and the peculiar dynamics such as beads-on-string patterns that these fluids exhibit in experiments, this paper presents a theoretical study of the dripping faucet for the case when the dripping liquid is non-Newtonian.

Although there are fundamental differences between the gross dynamics of breakup of liquid jets and liquid bridges and drops forming from tubes, they also exhibit many similarities on account of the analogous role that capillary pressure plays in each case as pinch-off nears. In a pioneering study, Goldin et al. [20] performed a linear stability analysis and carried out experiments to study breakup of viscoelastic jets. They showed that initially unstable disturbances grow faster on the surface of jets of viscoelastic fluids compared to that of Newtonian fluids having the same zero-shear-rate viscosity. Almost two decades later, Bousfield et al. [21] studied the nonlinear deformation and breakup of viscoelastic filaments by means of a numerical analysis of both the complete system of equations governing the problem and that of a one-dimensional (1D) thin filament approximation. They showed that while disturbances initially grow more rapidly on filaments of viscoelastic fluids than Newtonian ones, growth rate of disturbances at long times decrease for viscoelastic filaments relative to Newtonian ones on account of the large extensional stresses that develop in viscoelastic filaments. More recently, Brenn et al. [22] used linear stability analysis to study the onset of instability in jets of viscoelastic fluids moving through a surrounding gas whose dynamics they accounted for by treating it as an inviscid fluid. Moreover, these authors also compared their predictions with experimental results from the literature. Although these and other linear stability analyses are indispensable in developing insights into capillary thinning of non-Newtonian filaments, they cannot predict the dynamics close to pinch-off.

Two highly insightful papers, a theoretical one by Keller and Miksis [23], which proposed a scaling theory describing the self-similar recoil of an inviscid liquid sheet, and primarily an experimental one by Peregrine et al. [24], which suggested that the dynamics both temporally and spatially in the vicinity of breakup is universal, resulted in an explosive growth in interest in developing an understanding of interface pinch-off first in Newtonian fluids [25–33] and soon thereafter in non-Newtonian fluids [34–42]. Particularly noteworthy is the paper of Renardy [35] who used the 1D thin filament equations without inertia to study and contrast the asymptotic evolution and breakup of jets of Newtonian fluids and non-Newtonian fluids whose rheology are described by various viscoelastic constitutive equations. Among other things, Renardy [35] showed that a jet of an Oldroyd-B fluid does not break up in finite time. Renardy [35] further showed that a jet of Giesekus fluid, by contrast, does break up in finite time. Subsequently, Chang et al. [36] used both Oldroyd-B and FENE models and accounted for inertia to study the iterated stretching of viscoelastic jets.

Accompanying the recent flurry of activity in theoretical and computational studies of interface pinch-off, several researchers have recently begun to examine carefully in the laboratory breakup of jets and drops of non-Newtonian liquids. Mun et al. [43] experimentally studied the effects of polymer concentration and molecular weight on breakup of viscoelastic jets. To isolate the effects of extensional viscosity on jet breakup, they used fluids of constant shear viscosity, surface tension, and density. They found that fluids of low extensional viscosity had the same breakup lengths as Newtonian fluids of the same shear viscosity so long as the jet velocity was below a critical value. The breakup lengths of these viscoelastic jets became independent of velocity above this critical value. However, fluids of high extensional viscosity had breakup lengths that exceeded those of Newtonian fluids of the same shear viscosity. Christanti and Walker [44,45] experimentally studied breakup of viscoelastic jets subjected to natural and forced disturbances. They decoupled the effect of rate-thinning from that of rate-thickening by using fluids of constant shear viscosity but variable extensional viscosity. Christanti and Walker [45] showed that whereas formation of satellites can be suppressed using large disturbance amplitudes in jets of Newtonian fluids, satellites can be avoided altogether while using much smaller disturbance amplitudes by simply adding polymer to viscoelastic jets. These authors further showed that drop size distributions can be controlled by appropriately varying the molecular weight of the polymer. Cooper-White et al. [46] experimentally studied the dynamics of formation of drops of low-viscosity elastic fluids of constant shear viscosity and compared their behavior to drops of Newtonian glycerol–water solutions. They showed that during the initial stages of necking, inertia and capillarity control the dynamics, paralleling the response observed with Newtonian fluids [32,33]. These authors further showed that in such low-viscosity fluids, elastic effects come into play during only the final stages of breakup. In other words, these authors demonstrated that while the initial elongation of necked regions is dominated by the inertia of the drop, elasticity does strongly influence the resistance of the pinch regions to break-off. In a related study, Amarouchene et al. [47] carried out experiments that were similar to those of Cooper-White et al. [46] but used elastic fluids that rate-thinned to different extents and whose relaxation times were unspecified. Therefore, while Amarouchene et al. [47] reported the fascinating observation that the necks of such drops thin exponentially in time, they left unanswered whether elasticity alone was responsible for the novel thinning behavior.

Most studies on the deformation and breakup of non-Newtonian drops to date have focused on free drops suspended in another liquid and subjected to an external flow field [48–54] or atomization [6,55,56]. By contrast, our understanding of formation and dripping of drops of non-Newtonian liquids from a tube is weak due to the scarcity of experimental [46,47] and theoretical studies that have been carried out to date.

Heretofore most studies on drop formation have focused on deformation and breakup of single drops of Newtonian fluids. Some of these studies have been solely experimental in nature [57–59], solely computational in nature [60,61], and a combination of experiment and computation [31,33,62–64]. Although dynamics of single drops has received much attention for more than a century, the related problem of formation of many drops