Video-rheology – Studying the Dripping, Jetting, Breaking and “Gobbling” of Polymeric Liquid Threads

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ABSTRACT

In free-surface flows of low-viscosity complex fluids such as dilute polymer solutions and micellar fluids, viscous effects are frequently negligible. The dominant force balance may involve elastic, inertial, gravitational and capillary forces, and one is interested in the rarely-studied limit of \textit{inviscid viscoelastic fluid motion}. A quantitative analysis of such flows can be carried out using high-speed digital videoimaging. To illustrate this, we investigate thin jets of dilute polymer solutions at flow rates corresponding to a transition from dripping to jetting. The study reveals a novel periodic break-up process. In the initial stages, Rayleigh disturbances grow and the jet evolves towards the familiar beads-on-a-string structure. However the jet terminates in a large terminal drop which is almost stationary and appears to progressively swallow the beads on the string. As the mass of the terminal drop increases, it accelerates and ultimately pinches off. The process then repeats leading to a quasi-periodic break-up process. A simple axial momentum balance incorporating capillarity, gravity and fluid elasticity captures many of the essential features of this “gobbling” phenomenon.

INTRODUCTION

Unsteady jetting phenomena and film flows of low-viscosity complex liquids such as polymeric, surfactant and micellar solutions are ubiquitous in industrial operations such as inkjet printing and roll-coating. However the low viscosity of these fluids (typically 1–10 mPa.s) and the small relaxation times (typically \( O(\mu\text{ms}) \)) frequently makes it difficult to use conventional rheometric hardware to probe the viscoelastic properties of the materials. Furthermore the evolution of the fluid thread or film in the process of interest is frequently a ‘free-boundary’ problem; i.e. it is driven not by external boundary motion, but by forces such as gravity and surface tension.

In such cases, it makes sense to quantify the rheological response of the fluid directly in the process of interest, rather than strive to artificially constrain the motion to simple shear and pure extension. Such \textit{in-situ} analysis can be realized by combining high-speed digital video-imaging with automated image analysis and simple one dimensional balances of mass, momentum, and energy as appropriate. For brevity we refer to this combination of tools as video-rheology.

The ability to visualize and quantify fugacious phenomena using high-speed digital video-imaging has advanced tremendously in recent years due to increasing digital data-transfer, storage and processing possibilities. The slender geometry of pinching threads and films allows one to compare experimental observations with theoretical predictions, and also to analyze the dynamical processes in order to extract characteristic rheological properties of the complex fluids. Jetting transitions and break-up processes are examples of flows that can now easily be visualized and analyzed in this way. In this paper we will demonstrate the capabilities of video-rheology for analyzing uniaxial elongation in:

- elastocapillary filament break-up and propagation of recoil waves in threads of wormlike micellar solutions;
- Periodic and quasi-periodic drop dynamics that arise in dripping and jetting of micellar and polymeric solutions expelled from nozzles of different diameters.

In the latter example, analysis of the jet dynamics in the flow transition region between dripping and jetting reveals the existence of quasi-periodic oscillations in the size of the drops formed. This is in sharp contrast to the regular periodic pinching of small drops that is observed in simple Newtonian fluids. As a result of fluid elasticity the ultimate pendant drop remains attached to the primary fluid jet in a ‘beads-on-a-string’ configuration and ‘feeds’ on the growing secondary droplets until it exceeds a critical size, at which point it detaches and the process repeats. An example of this gobbling phenomenon is shown in Figure 1. An axial momentum balance
incorporating capillarity, gravity and fluid elasticity captures many of the essential features of this “gobbling” phenomenon.

**EXPERIMENTAL DATA ANALYSIS**

Experiments have been performed with a number of dilute polymer and surfactant solutions. The results reported below relate to 100 ppm solution of a linear partially hydrolyzed polyacrylamide (Praestol 2540, Stockhausen; molar mass \(M_w = 14.0 \times 10^6 \) g/mol) in deionized water. The test fluid is in the dilute regime with \(c/c^* = 0.75\). The zero shear rate viscosity and the surface tension for this solution are \(\eta_0 = 2.74\) mPa-s, and \(\gamma = 61.4\) mN/m. The longest relaxation time \(\lambda\) was determined via capillary thinning experiments carried out using a CABER-1 extensional rheometer (Cambridge Polymer Group) and was found to be \(\lambda \approx 0.012\) s.

Thin jets were expelled vertically downward or horizontally from standard nozzles of different inner diameters in the range \(0.10 \leq 2R, \leq 1.52\) mm. The jet breakup was monitored using a Phantom high-speed digital \((1K \times 1K)\) camera with frame rates up to 2400 frame/s. First, the critical flow rates corresponding to transition from jetting to dripping were determined for water and polymer solutions. The “jetting” transition in the case of water was defined as the flow rate at which a continuous jet of relative length \(L/R >> 10\) existed. Results are shown in Figure 2. These data are in reasonable agreement with theory [1] (solid line in Figure 2). The critical flow rates of the jetting transition for polymer solutions are much smaller than the corresponding values for water (cf. Figure 2). For dilute polymer solutions, the size of the drops developing in the course of jet breakup grows dramatically as the flow rate approaches the critical value. This growth occurs in a narrow range of flow rates above the critical one, and is rather sensitive to specific operating conditions.

**RESULTS AND ANALYSIS**

Detailed analysis of the gobbling phenomenon was performed using frame-by-frame computer processing of digital images of the jet. The familiar beads-on-string structure that develops on the jet prior to breakup, allows us to use small drops as markers, which are clearly distinct from large terminal drop. In this way, ‘time-space’ diagrams were constructed (see Figure 3 overleaf). These \(XLT\) plots show the positions of the terminal drop and the evolving “beads” at different times.

Analysis and processing of \(XLT\)-diagrams reveals that in the ‘gobbling regime’ characteristic of thin polymer jets, the dynamics of gobbling is primarily controlled by the momentum balance of the terminal drop, and that the net force exerted on the drop by the jet can be closely approximated by \(\pi \gamma R\). The primary effect of polymer additives consists in preventing premature breakup of the liquid filaments between consecutive beads in the jet.

The critical condition for existence of a continuous jet is that the momentum flux in the jet is positive. This criterion results in a surprisingly reasonable prediction of the critical flow rate (see Figure 2) by a simple formula:

\[
Q_{cr} = Q_{cr0}\left(1 - \frac{\rho g LR}{\gamma}\right)^{1/2}; Q_{cr0} = \frac{\pi \gamma R^{1/2} (\gamma/\rho g)^{1/2}}{2}
\]

where \(L\) and \(R\) are the jet length and radius respectively.

The same momentum balance implies that during each cycle the terminal drop (of mass \(M(t)\)) moves according to the parabolic law:

\[
L(t) = (V^* - U)t - at^2; M = \rho RT^2(V^*t - at^2)
\]

where the ‘effective acceleration’ is \(a = g/6\).
The derivation of this expression follows that in [1], the only difference is that here \( V^* = (\gamma \rho R)^{1/2} \). Figure 4 shows a comparison of these expressions with experimental data derived from Figure 3.

![Figure 3. XLT-diagram of the gobbling phenomenon. Positions of the terminal drop and ‘marker’ beads are shown by blue and red dots respectively.](image)

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The expressions (2) show that the force exerted on the last drop is \( F = M(g - 2a) \approx (2/3)Mg \). In the last ligament this force is primarily supported by elastic stress. However, it remains almost constant along the continuous part of the jet, and therefore the effective stress can be estimated through the upstream value resulting from surface tension as \( F = \pi \gamma R \). The maximum mass of the terminal drop is then estimated as \( M = (3/2)F/g = 3\pi \gamma R/(2g) \). A mass balance also yields \( M = \rho QT \), the oscillation period is then

\[
T \approx \left( \frac{3\pi \gamma R}{2g} \right) \sqrt{\frac{Q_{cr} \rho}{\rho}} = 3(\gamma \rho R)^{1/2}/(2g) \tag{3}
\]

These expressions are estimates based on the simple ‘parabolic law’ solution. A more detailed dynamical theory predicts reasonably well the near-critical oscillatory dynamics, provided that the critical jet length at breakup is set to be equal to the experimental one. At leading order the presence of polymer does not significantly affect the critical flow rate, maximum end drop size or the oscillation period. Its primary role (at least, in the ‘inviscid elastic’ regime) is to set the timescale of the drop detachment or \( L_{br} \).

The polymer molecules eventually approach limiting extension and then the filament breaks in much the same manner as in a Capillary Breakup Rheometer, albeit with different kinetics due to existence of the non-vanishing axial stress.

A simple theory of filament extension by a constant force \( F \) shows that for large enough polymer extensibility there are three well-defined stages of extension, namely a short initial viscoelastic stage, a long quasi-equilibrium stage when the filament radius decreases exponentially, \( R(t)/R_0 \approx (G \pi R_0^2 / F)^{1/2} \exp(-t/2\lambda) \) followed by a short quasi-viscous breakup stage, when the effects of finite extensibility become important.

We use this theory to predict the breakup time \( t_{br} \) for ligaments in the beads-on-string structure of jets of different initial diameter. These calculations were performed assuming a limiting FENE extensibility parameter \( b = 6.1 \times 10^4 \) computed from the known molecular weight of the PAA molecules. The critical jet length is such that after the fluid particle flight time \( t_{br} \), the last ligament breaks, and the terminal drop detaches. In Figure 5 we show experimental data on the critical flight time (scaled with the relaxation time) and the numerically-predicted values of the ligament breakup time:

![Figure 5. Breakup time vs. initial nozzle radius \( R_0 \). circles: experimental data; broken line: linear regression; solid lines: from FENE-P calculations.](image)

Figure 5. Breakup time vs. initial nozzle radius \( R_0 \). circles: experimental data; broken line: linear regression; solid lines: from FENE-P calculations.

**CONCLUSIONS**

By using a combination of detailed image analysis with elementary one-dimensional balance relations we have been able to understand the gobbling phenomenon as essentially a “delayed dripping”, and describe the spatial and temporal dynamics in quantitative terms. The main role of the polymer is to delay jet breakup and hinder the jet re-attachment to the nozzle. Consequently a much larger terminal drop is created.

**REFERENCES**